Anonymous Referee #1
Received and published: 30 April 2013
The authors modeled the emission and global transport and deposition of radioactive Cs from the Chernobyl nuclear power plant accident using the LMDzORINCA model at different resolutions. Concentrations and deposition quantities were compared to measurements and other studies in the literature. This type of study is certainly of value in the context of the simulation of the atmospheric transport of radionuclides.

The reviewer recommends publishing this paper with major revisions in response to the following questions and comments.

General Comments:
Sec. 4: Although it's important to provide the technical specifications, too much detail, all available in other publications, not unique to this study and unnecessary for the scope of this paper is provided on the parameterisation of deposition processes in the model. This section serves as a digression and should be shortened and simplified.
Response: We agree with the reviewer. Therefore, we have transferred the chapter to the Supplementary Material-Methodology.

The treatment of statistics in Sec. 5 needs to be improved. For this point, also see individual comments below.

Individual Comments:
Please find below individual comments prefixed by page and line number.

p7687 l1-3: It's not clear in the text that the model was nudged for this study. Please add additional details: What reanalysis data was used (eg. ERA-40), time constant?
Response: Corrected (P5-L10 MS with track changes)

p7687 l8: Please clarify in the text how Cs137 is treated in the model – "mostly" here is ambiguous. Also, if no gas phase chemistry is included in your simulation, sentence on line 6 p.7686 should be removed as it's unnecessary and may confuse the reader.
Response: We state in Page 6 – lines 12-13 that Cs137 is treated as an aerosol tracer. We remove the sentence on gas face chemistry (see manuscript in track changes mode)

p7687 l26-27: No need to quote each day and percentage. Just refer to Table 1 to improve legibility. Table 1: You refer to Devell et al, 2002 in the caption; yet only Devell et al. 1996 appears in the reference list. Further, the 1996 publication does not include the day-by-day or vertical profile for the emissions. Please provide correct references.
Response: The reference in the caption of Table 1 has been changed to Brandt et al. (2002). Percentage values in the "Emission estimates after the accident" chapter have been removed (p.7-L.32 –track change ms).

p7688 l21: Particle size distribution functional form should be added to the text for completeness. Fig. 6: What are the R^2 values? No description is given in the caption or the text.
Response: Corrected (P2-L8 in Supplementary information now). R^2 is now explained in the caption (p.31-L29)

p7693 l13:"altitude of the source" -> number of emission vertical levels; spread of emissions was greater -> emission distribution had more points; layers were denser covering lower distances-> layers were separated by shorter distances
Response: Corrected according to the reviewer's suggestions (p.9-L. 6 and 7).

p7695 l8,9: From Fig. 7 the isosurface does not "dominate the higher layers across all Europe". For example, nothing appears aboveWestern Europe. Also, the 19-layers run rises to a higher altitude (lower pressure) than the 39-layers in the figure, in contrast to what is claimed in the text l.14,15. If this is not the case, both panels should be plotted against the same scale in the vertical for the panels to be comparable. A vantage point more similar to Brandt et al. is also needed to help facilitate the comparison. Overall, I don't think that as it is Fig. 7 contributes much to the paper and it should be improved or removed.
Response: Obviously the 2 figures of different vertical levels had been placed in an opposite way. They are now correct and what is mentioned in the manuscript conforms to fig. 7. We appreciate the reviewer for his comment.

p7696 l8: What does "averaging" refer to? Should be clarified in the text.
Response: Sentence has been removed, as it does not enforce the statements of the paragraph (p11-L.16, track change ms).

p7696 l14: "determination recoveries contrast between methodologies" is not clear.
Response: In order the sentence to be clearer, we link it with the previous sentences, which explain why the different methodologies used for the construction of the Atlas map might be responsible for some discrepancies between model and measurements (p.11-L.21 track changes ms).

Figs 8-10 captions: Website should be moved to references. Location (North Europe, etc.) should be moved to the first line. You mention "north, west, south-eastern", yet present "central-western, north, south-eastern". Sentence beginning "They were examined according..." appears in the text and it's superfluous to be included in each caption.
Response: These figures have been moved to the Supplementary Material of this manuscript in order to limit the manuscript in a way to be more easy-to-read for the readers.

p7697 l.5: "Educated guesses" needs to be elaborated upon. How were they calculated?
Response: The first estimation of the source term was based on a USSR report to the International Atomic Energy Agency (IAEA) in 1986 (Persson et al., 1986; Hass et al., 1990), where the source was estimated on the basis of summation of the material deposited within the countries of the former USSR. These investigations did not take into account the material deposited outside the former USSR and has since been corrected several times from other investigations with more than a factor of two. The emissions used in the simulations (see Table 1) were taken from Brandt et al. (2002) and they are based on Waight et al., 1995. The amount of release and temporal variation used in this study is similar to the estimates in De Cort et al. (1998).
Here, we state that they are "educated guesses" because (a) an uncertainty of at least ±50% is used, and (b) the altitude of the emissions (which is very important and can change the transport regime extremely, as shown in the RG19L(S) simulation) is based

on simple assessments. The last is mainly because tools such as back trajectories were not available in 27 years ago, and also, there was lack of information, while the national monitoring systems of the countries were not that developed like nowadays.

p7697 l.22: What do you mean by "the correlation coefficient at 95% confidence level"? A p-value needs to be computed for the test to decide significance at 95%.
Response: I agree with this statement. Here we mean that p was always lower than 0.05! It is now mentioned in the captions of Tables 3 and 4 and in p16-L11, 13, in p.12-L23, in p.13-L.4.

p7698 l.5: See previous comment on statistical significance.
Response: Clarified in the text. We appreciate the reviewer for this comment..

p7967 l.25: 0.81 does not appear anywhere in Table 3. What do you mean by "real emission altitude", when you also refer to "the emission altitude was taken into account" in l.22? The text needs to be clarified.
Response: The right value is 0.84 and is now included in the text. In the text we follow the pattern of (a) assuming surface emissions, and (b) real emission altitude, which means all the other simulations with the tracer emitted in a certain altitude (according to Brandt et al., 2002). Besides, we define in p.7-L.17 that "real emission altitude" means the simulations where emissions according to Table 1 have been taken into account.

p7699 l.1, Fig.12: It's my understanding that the Pearson's linear relation coefficient indicated the strength of the linear relationship but says nothing about the slope. Good agreement can be claimed if the lines fall close to the 1:1 slope. What is the case here?
Response: The Pearson test is the simplest statistical test and it is frequently used when similar quantities are compared. It is a measure of the linear correlation (dependence) between two variables X and Y, giving a value between +1 and −1 inclusive. It is widely used in the sciences as a measure of the strength of linear dependence between two variables. In our case where modeled and measured quantities are compared, the unique +1 dependence would follow the function y=x, whereas for -1, would give a reversely proportion dependence (completely wrong here!!!). The slope would give a glance of what is overestimated or underestimated against what, which it can be seen very easily in the relevant figure.

Fig. 20 caption: "Linear fitting" here refers to the 1:1 line? Should be made clear.
Response: Yes, it refers to 1:1 dependence and it has been clarified now in the text (p16-L.32).

Technical Corrections:

p7683 l9: be -> by
Response: Corrected (p.3-L.6)

p7686 l15: plane -> dimension
Response: Corrected (p.5-L.23).

Sec 5.1 title: versions -> resolutions
Response: Here, we have decided to maintain the term. For example, it refers to the

zoom version of the model, which uses the same number of points in longitude and latitude as in the regular grid, but it stretches the grid over specific regions (as seen in Fig. 1)

Figs. 2-5: The captions read that every 10 days in May are shown, yet only one plot appears for May in each.
Response: Corrected (Fig.2-5)

p7697 l.23: confident -> confidence
Response: Corrected (Tables 3 and 4 and in p16-L11, 13, in p.12-L23, in p.13-L.4)

p7699 l.23: Remove "consequently" p7699 l.25: Remove "briefly"
Response: Corrected (p.14—L.16 L.18)

p7700 l.1: appeared to be a local event -> was limited locally.
Response: Corrected (p.14—L.21)

p7700 l.25: is not able to estimate -> underestimates
Response: Corrected (p.15-L.11)

p7701 l.10: Remove "where"
Response: Corrected (p.15-L.25).

p7703 l.1: deficiencies -> discrepancies
Response: Corrected (p.17 – L.8).

The emission inventory used in the study (Devell et al.?) should be included in the abstract.
Response: Corrected (p.2-L.13)

Abstract l11: "The best choice for the model validation was the"-> "The model is validated for the"
Response: Corrected (p.2-L.12)

Abstract l12: Second sentence: no need for "However,"
Response: Corrected (p.2-L.20)

Abstract l24: "Atlas" here is ambiguous. Please add better description or reference
Response: Corrected (p.2-L.22)

Anonymous Referee #2
Received and published: 29 May 2013
The authors present the simulations of the transport, wet and dry deposition of the Cs-137 released during the Chernobyl accident. The simulations were carried out with the coupled model LMDzORINCA at the European scale. Several configurations of the model were studied. Results were compared to the REM database and to other studies already published. The paper is interesting and addresses important questions to model the atmospheric dispersion of an accidental release. The reviewer recommends its

publication after improvement.

GENERAL COMMENTS

- To compute the wet deposition, it is essential to have realistic precipitation fields. The quality of the LMDz precipitations and more generally, the differences between the LMDz fields compared to the ERA-40 fields should be discussed. Does the vertical resolution of the simulations impact the precipitation fields (especially the convective precipitation fields) / the scavenging height?

Response: Below, we attach precipitation fields from observations and the model. In the first one, the average precipitation in mm/d is shown for ERA40 (http://data-portal.ecmwf.int/data/d/era40_daily/) precipitation fields (2.5x2.5 degrees) for the year 1986. In the second picture, the average precipitation (in mm/d) from our model (for 1986) is shown for a horizontal resolution of 0.66x0.51 degrees.

As you can see the difference in Europe (inside the zoom area) is very small! We have included relevant comments in the manuscript (Chapter 4.4). The average relative discrepancy (percentage) between model and observations ([mod-obs]/obs) is 8% in a box of 700x700 km and reaches 10% in 3000x3000 km centered in the plant. The difference was estimated after re-gridding ERA40 (2.5x2.5 degrees) to LMDZ resolution (0.66x0.51 degrees).

The vertical distribution of scavenging is determined by the height of which precipitation is defined. In LMDZORINCA we used Emanuel's scheme (see supplementary materials) for convection, which has been defined in the 1-D column of the model, by comparing cloud properties and precipitation from experiments in mid-latitudes and tropics.

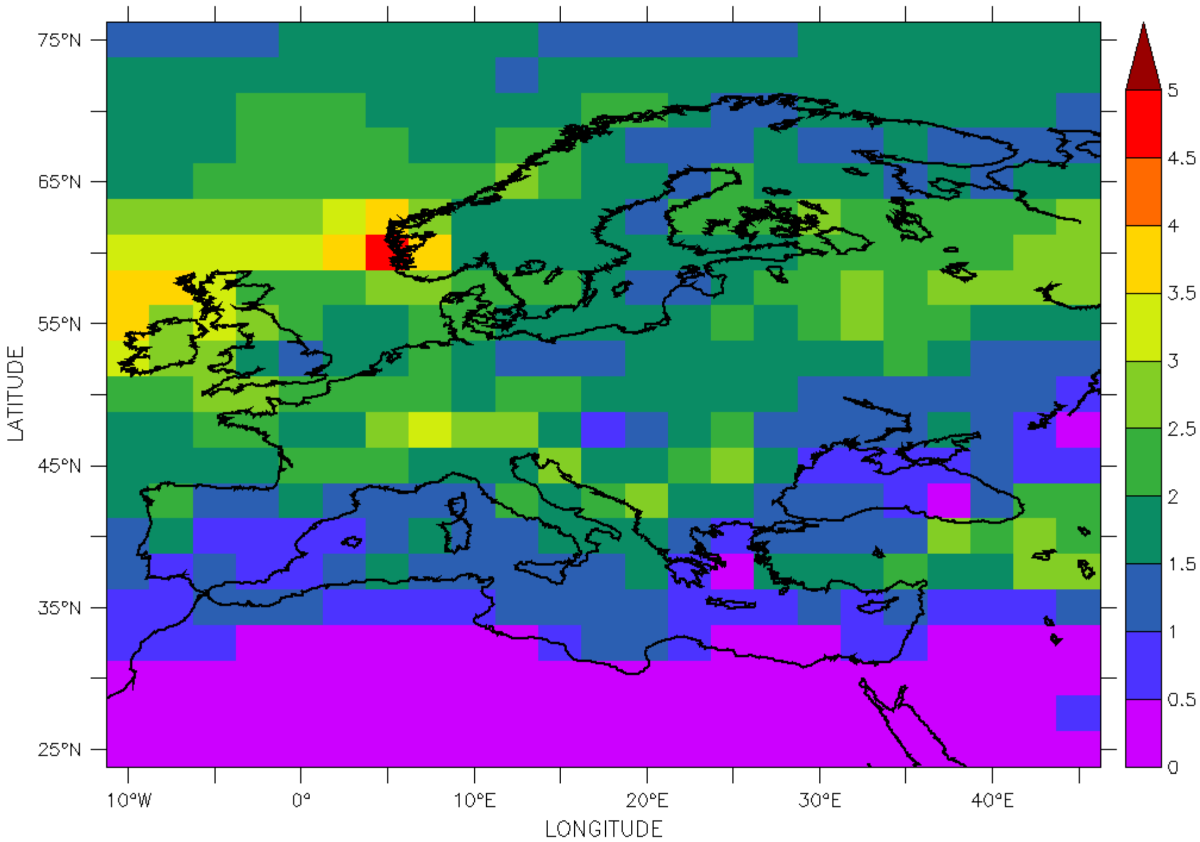

Figure 1. ERA40 (2.5x2.5 degrees) average precipitation (in mm/d) over Europe in 1986.

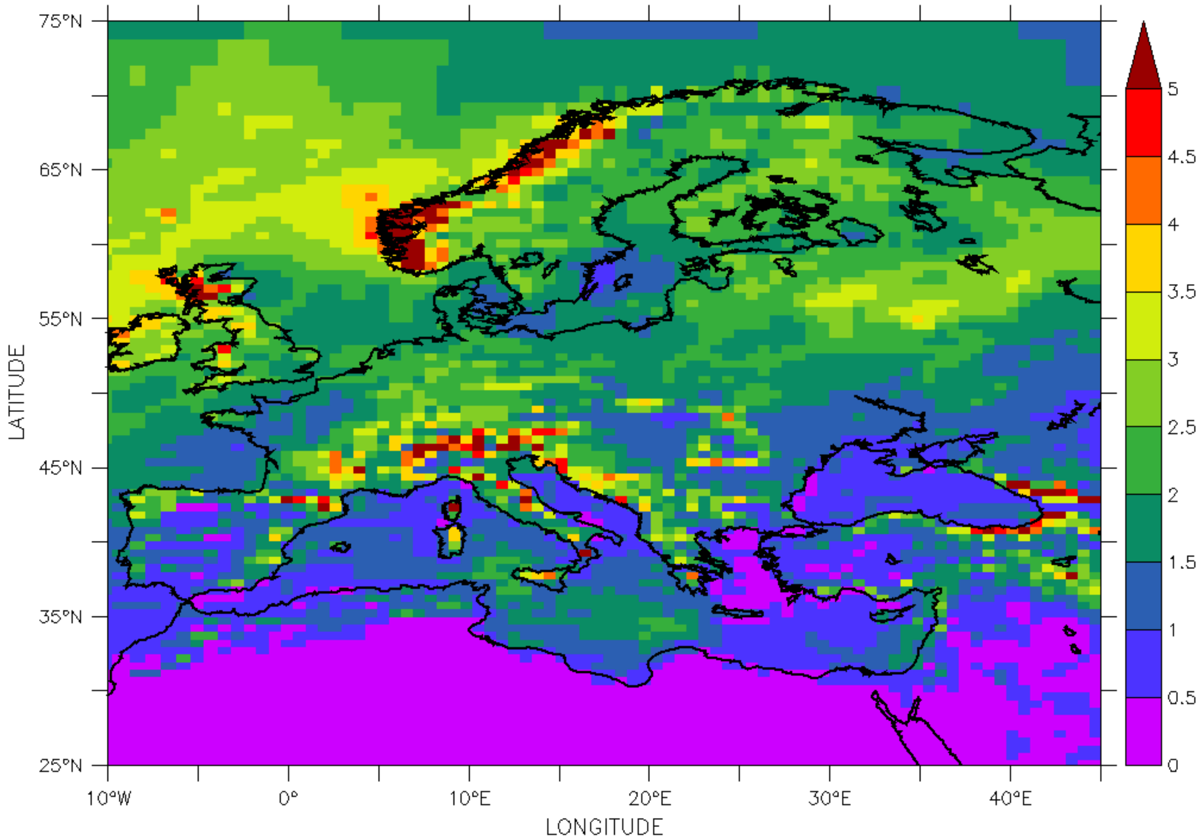

Figure 2. LMDZ (0.66x0.51 degrees) average precipitation (in mm/d) over Europe in 1986.

- The uncertainties on the precipitation fields should be one of the reasons discussed in chapters 5.3 and 5.4 to explain the discrepancies between the observed and modeled deposition.

Response: Below, we compare precipitation fields from the model and observations (2 attached pictures). In the first one, the average difference of precipitation in mm/d is shown between LMDZ model and ERA40 (http://data-portal.ecmwf.int/data/d/era40_daily/) precipitation fields (regridded from 2.5x2.5 degrees to the grid of LMDZ) for the year 1986. In the second picture, we calculate the same difference between our model (for 1986) using the annual average precipitation fields from the Global Precipitation Climatology Project (GPCC, http://www.esrl.noaa.gov/psd/data/gridded/data.gpcc.html) (in mm/day) for the period 1981-2010 (regridded from 0.5x0.5 degrees to the grid of LMDZ). As we explained before the relative difference in our region of interest is small and this is also show below in mm/d.

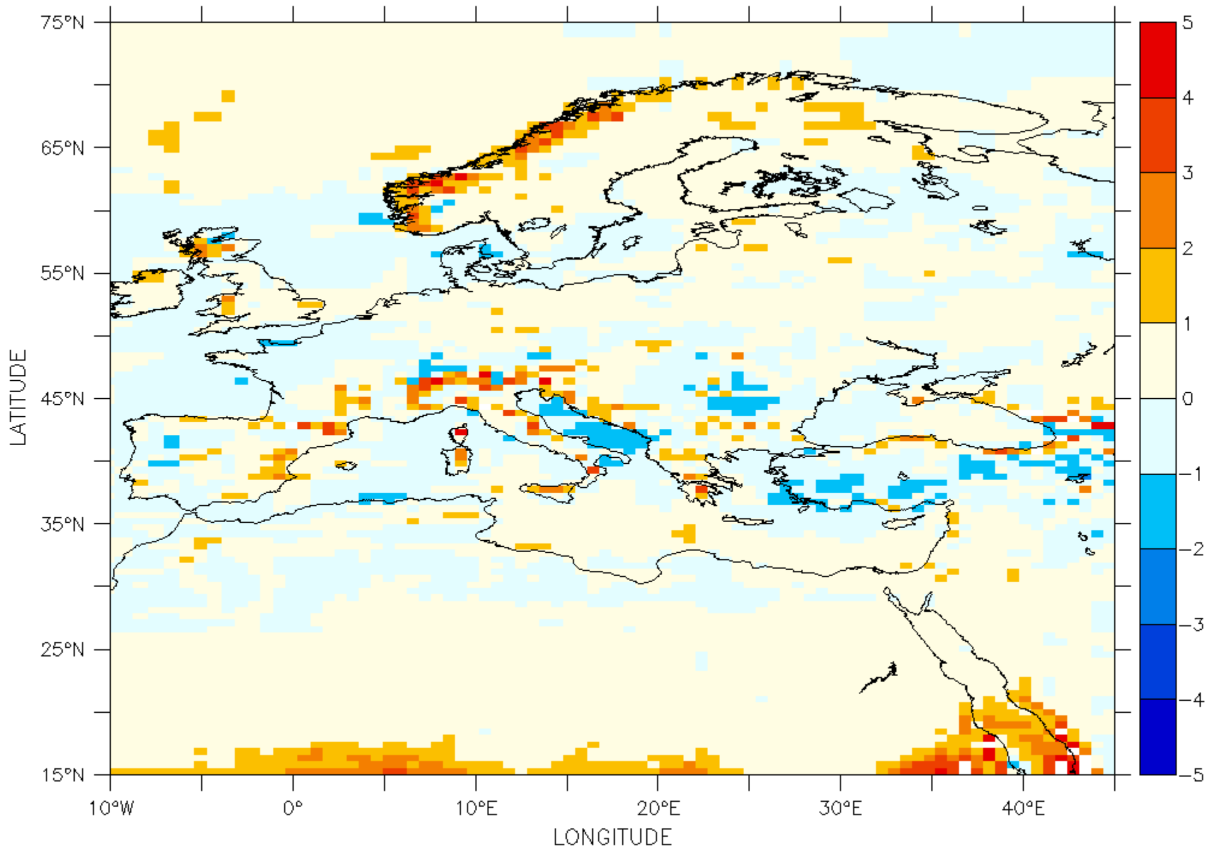

Figure 3. Average difference of precipitation in mm/d is shown between LMDZ model and ERA40 for 1986.

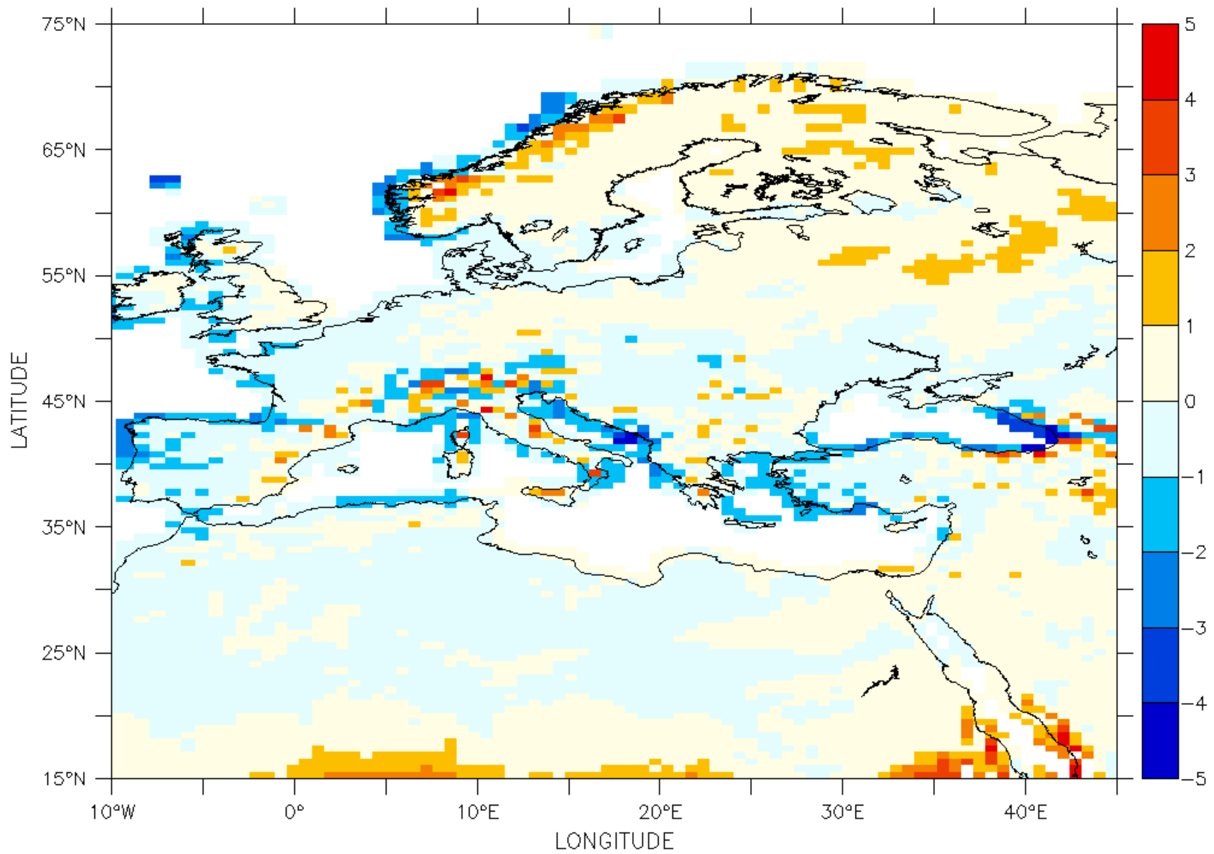

Figure 4. Average difference of precipitation in mm/d is shown between LMDZ model and GPCC.

- The authors have to precise which parameterization they use for the horizontal and vertical diffusion processes. Do the vertical and horizontal resolutions impact the Cs-137 dilution? Does the choice of the parameterization for the diffusion may explain the differences between the simulations done with the different resolution?
Response: Paragraphs for the parameterization of diffusion and convection have been added (P4-L12 at the Supplementary Material of this article).

- Chapter 4: Why the wet deposition is parameterized assuming the Cs-137 behaves as a soluble gas and not as a particle? The particle size should influence the wet deposition.
Response: Our sentence was inaccurate and has been corrected. We thank the reviewer for pointing it out. We wanted to indicate that in-cloud scavenging of Cs was treated as for a soluble gas. In addition we account in the code for below-cloud scavenging and sedimentation of 137Cs. Hence 137Cs is treated as a sub-micronic aerosol.

- Chapter 5: The text should be improved:
o Information is repeated.
Response: The manuscript has been significantly improved in order to be more precise and easy-to-read for the reader. Many parts and figures have now been moved to the Supplementary Materials and the most important parts have been maintained.

o The impact of the release height is too highlighted compared to the impact of the horizontal and vertical resolution. It is true that the influence of the release height is very important but the test of only 2 release heights so different is a bit extreme.
Response: We agree with this comment. The idea was not to simply to compare 2 extreme heights of emission but 2 real facts: Wrong information (which forces the use of surface emissions) versus educated guesses (that almost all the scientists that simulated the accident before have used). However, the most important part of this analysis is the consequences if decision-making has to be done after a major event like Chernobyl, which may save thousands of lives. This is the example we give for the city of Kiev.

o You should give the fac2, fac5... scores to be able to better compare your results with those of Brandt et al., Quelo et al., 2007. . .
Response: The actual goal here is not to compare our results with those of Brandt et al. but with the REM database. We use 3 statistical tests (Pearson, Spearmann, Kendal's Tau) and we also calculate biases in order to find the discrepancy from the measurement. These biases are discusses based on what Brandt et al. found instead. Insisting in a comparison with Brandt's paper would mean that we try to compete on the better response of the model, which is not the case in this paper. We have clarified the lines.

o The statistical analysis should be improved.
Response: We give 4 different statistical metrics (Pearson, Kendal Tau, Spearman correlation coefficient and the relevant calculated biases). We believe they are enough to prove that the model produces reliable results. However, if the reviewer has to suggest anything more specific, we would be willing to make further changes

INDIVIDUAL COMMENTS
- The organization of the introduction should be improved in order to highlight the

objectives of the study
Response: In Chapter 1 (introduction) we indicate the major findings reported after the accident by several scientists worldwide in terms of emissions and consequences. The last paragraph of the chapter has been used to highlight the objectives of the study in order to be clear to the readers. We do not know what the reviewer suggests here.

- P7683 the sentence L9 “the absence of reliable. . .” should be clarified.
Response: Here, the expression “lack of reliable information” has been used to comment what really happened in Europe after the accident. Other countries were commenting the detected radionuclide concentrations (although most of them did not have organized monitoring stations) and other countries were claiming that they did not detect the radioactive cloud at all (although it proved they did years after), in order not to panic the population. We now put a comment on that in the manuscript.

- P7685: “the already known patterns of the releases” is too strong as explain later the releases are highly uncertain.
Response: We changed the expression to “reported patterns of the releases”

- P7685 the last sentence is not useful.
Response: We understand the sense of the reviewer’s comment, and we have removed the last sentence.

- P7684 last paragraph: many studies have been performed in order to validate long range dispersion models with the Chernobyl accident: the Brand’s PhD work, the Quelo et al. 2007 (Atm. Env.) paper.
Response: The reference of Quelo et al. (2007) has been also included in the reference list.

- Chapter 2: you need to indicate the met data you use to nudge LMDz and the temporal resolution.
Response: Corrected (Page 6-L.10 in the MS with track changes)

- P7687 L10: the references should be ordered according to the year of the paper.
Response: Corrected

-Chapter 4: which aerosol distribution do you use?
Response: Corrected (P2-L8 in Supplementary information now).

- P7691 first paragraph: you should compare your deposition velocities to the deposition velocities given in the Sportisse paper which are usually used to model the deposition of radioactive materials.
Response: This part has been now put to Supplementary Material of this paper. We have included the values of the deposition velocities from the paper according to the reviewer’s comment. We appreciate for his help.

- Chapter 5: you use fallout to refer to the plume. Fallout is ambiguous since it is often used for the deposition. You should use “the plume” instead.
Response: We do not agree with this comment. I provide the official explanation as presented in several online dictionaries: “The slow descent of minute particles of debris

in the atmosphere following an explosion, especially the descent of radioactive debris after a nuclear explosion". Since we talk about Cs-137, which is a particle, we believe it fits very well. However, if it still causes a problem, we would be willing to change the word in a next step of the reviewing process.

- Chapter 5.1: you should better highlight the similarity and the discrepancies between the different simulations.
Response: Some changes on this direction have been done on the manuscript. The description of the runs performed is being presented now in Chapter 4, where the main similarities observed are presented. If something more specific is to be changed, we would will to correct it.

- P7691 the sentence L9-10 is too reductive and unnecessary.
Response: Corrected according to the reviewer's comment.

- P7693 L25: "the cyclone observed . . .": was the cyclone really observed? Why it is not discussed before with the other simulations. Was it simulated?
Response: The cyclone was observed by the step-by-step transport of the Cs-137 plume and can be seen in the 2d-movie in the Supplementary Materials of this article. It is not discussed further because, in our opinion, it would not give any extra benefit to our discussion.

- P7695 first paragraph: the description of the 3-D illustration is not clear enough. I do not see the benefit. You should improve the analysis of the figure and try to give some possible explanations for the differences.
Response: We have changed this figure after determining some crucial mistakes. Also some parts in the manuscript that analyse it. We appreciate the reviewer for his comment.

- Chapter 5.2: You should improve the statistical analysis and your conclusions.
Response: This is too obscured. We have validated the comparison using 2 different statistical tests and also calculated biases in the relevant figures. We could do further changes if something more specific is asked.

- P7698 Add X, Y meaning.
Response: X and Y here represent the variables being compared (model versus modeling). We have added a comment that.

- Chapter 5.3 P7700 L9-12 "It is unexpected. . ." You should remove this sentence or explain more precisely why you say that. What is usually done for crisis management?
Response: We do not understand the problem here. We provide an example of how the lack of available information after that major accident could have caused problems after using such modeling tools to predict the areas of the highest contamination. The official authorities were claiming that there was not any explosion in the beginning. Using this assumption we emit Cs-137 from the surface. And here comes the example of the Kiev city. According to our hypothetical simulation and taking into account the information of the first days following the accident, we estimate that there would be extreme contamination of the city that would lead to evacuation activities. Anyway, the paragraph has been changed in order to be more comprehensive.

- P7703 First paragraph: Do the other models have the same bias with the measurements? I have the feeling that the parameterization of the deposition and especially errors in the precipitation fields may explain those biases?
Response: To our knowledge model comparisons with REM database present similar to our biases. We provide evidence in some previous comment-responses why there is no problem with the parameterization of deposition or the precipitation fields used.

- Conclusions P7705: "e.g. using inverse modeling" you should add some references.
Response: Reference has been added. We thank the reviewer.

- P7705 "knowing the exact core. . ." I do not see why you add this point. It would be better to discuss what could be done to improve the scores of the model-to-data comparisons.
Response: We try to find the source of these discrepancies. Having seen what the other models find for the Chernobyl accident, we believe that our results can be considered as very convenient. We believe that the 2 main sources of these discrepancies are (a) the source inventory (that it is known with a 50% uncertainty) and (b) the exact altitude that the emission took place (in this point, there are only guesses and rough estimations, although previous modelers have used the same estimations). This is exactly what we discuss in the manuscript.

- Table1: you should give the layer thickness.
Response: Corrected (Table 1) according to the reviewer's suggestion.

- Fig.7: You should plot the Brandt et al. Figure to help for the comparison. You should add a vertical scale.
Response: Obviously, Brandt et al. have used different software to create the 3d representation of the iso-surfaces. In both cases the altitude scale is not given. Therefore, we do not understand how it would help to "copy a figure on another". If the reviewer has something more specific to suggest, we would be glad to follow it in a next step.

Simulations of the transport and deposition of ^{137}Cs over Europe after the Chernobyl NPP accident: Influence of varying emission-altitude and model horizontal and vertical resolution

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Abstract

The coupled model LMDZORINCA has been used to simulate the transport, wet and dry deposition of the radioactive tracer ^{137}Cs after accidental releases. For that reason, two horizontal resolutions were deployed and used in the model, a regular grid of $2.5^{o}\times1.27^{o}$, and the same grid stretched over Europe to reach a resolution of $0.66^{o}\times0.51^{o}$. The vertical dimension is represented with two different resolutions, 19 and 39 levels respectively, extending up to mesopause. Four different simulations are presented in this work; the first uses the regular grid over 19 vertical levels assuming that the emissions took place at the surface (RG19L(S)), the second also uses the regular grid over 19 vertical levels but realistic source injection heights (RG19L); in the third resolution the grid is regular and the vertical resolution 39 vertical levels (RG39L) and finally, it is extended to the stretched grid with 19 vertical levels (Z19L). The model is validated the Chernobyl accident which occurred in Ukraine (ex-USSR) on May 26th 1986 using the emission inventory from Brandt et al. (2002). This accident has been widely studied since 1986, and a large database has been created containing measurements of atmospheric activity concentration and total cumulative deposition for ^{137}Cs from most of the European countries.

According to the results, the performance of the model to predict the transport and deposition of the radioactive tracer was efficient and accurate presenting low biases in activity concentrations and deposition inventories, despite the large uncertainties on the intensity of the source released. The best agreement with observations was obtained using the highest horizontal resolution of the model (Z19L run). The model managed to predict the radioactive contamination in most of the European regions (similar to De Cort et al., 1998), and also the arrival times of the radioactive fallout. As regards to the vertical resolution, the largest biases were obtained for the 39 layers run due to the increase of the levels in conjunction with the uncertainty of the source term. Moreover, the ecological half-life of ^{137}Cs in the atmosphere after the accident ranged between 6 and 9 days, which is in good accordance to what previously reported and in the same range with the recent accident in Japan. The high response of LMDZORINCA model for ^{137}Cs reinforces the importance of atmospheric modeling in emergency cases to gather information for protecting the population from the adverse effects of radiation.

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1. Introduction

The Chernobyl Nuclear Power Plant (NPP) accident on 26 April 1986 resulted in the dispersion and deposition of a large amount of radionuclides into the environment. On April 26th 1986, two explosions took place in the power plant releasing and transporting radioactive materials over long distances. The absence of reliable models in the period of the accident and the lack of reliable information (e.g. absence national well-organized monitoring centers providing support to the official authorities) on the direction taken by the released elements motivated several researchers to develop environmental modeling tools, in order to be able to study potential accidental scenarios. Since then, many national and international efforts have been initiated to develop reliable models that will be able to describe transport and dispersion mechanisms when large amounts of radionuclides are released. Such tracer models can be used to estimate the spatiotemporal distribution of the fallout from accidental releases and the output can be used for preventive purposes, as well as to estimate the exposure and the harmful impacts from the dangerous compounds on humans, animals and vegetation.

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It has been estimated that over 10 EBq ($\times 10^{18}$ Bq) of fission and activation products escaped from the damaged reactor (De Cort et al., 1998), whereas 2 EBq (the most refractory) were deposited in the 30 km vicinity of the power plant (Hatano et al., 1998). The most abundant nuclides were ^{133}Xe (~ 6500 PBq), ^{131}I (1200 – 1700 PBq), ^{132}Te (1000 – 1200 PBq) ^{137}Cs (~85 PBq), ^{90}Sr (81 PBq), ^{134}Cs (44 – 48 PBq), whereas the most refractory, less volatile radionuclides were ^{144}Ce, ^{141}Ce, ^{106}Ru, ^{140}Ba, ^{95}Zr, ^{99}Mo, $^{238-241}$Pu etc... (Devell et al., 1996; De Cort et al., 1998). However, a radionuclide of major concern is ^{137}Cs, due to its half-life (30.2 y), the radiation type it emits during its radioactive decay and its bioaccumulation by organisms. Consequently, it is a chemical analogue of potassium and rubidium with high mobility in biological systems. Its chemical and metabolic-physiological reactions are similar to those of potassium (Woodhead, 1973) that is essential for many organisms. This explains why ^{137}Cs gets enriched within tissues and cells. However, Cs cannot easily replace K in its metabolic functions, and it is not usually received by organisms in the same portion as potassium (Kornberg, 1961). Finally, it also participates in the augmentation of the total radioactivity to which the population is exposed.

Despite the dramatic consequences of the Chernobyl reactor accident, the atmospheric releases and the observed deposition of radionuclides provide a challenge for the modelers to test and improve their long-range dispersion models. For many years, operational codes have been developed to quantify the global fluxes of chemical pollutants (Elliassen, 1978; Elliassen and Saltbones, 1983; Prather et al., 1987; Lee et al., 2001; Stier et al., 2005; Koch et al., 2009; Huneeus et al., 2011; Olivié et al., 2012 and many others). At the same time, some authors proposed the use of certain codes to analyse and/or predict the atmospheric transfer of radionuclides (ApSimon et al., 1985; ApSimon et al., 1987; Jacob et al., 1987; Albergel et al., 1988; Lange et al., 1988; Hass et al., 1990; Piedelievre et al., 1990; Balkanski et al., 1992; Klug et al., 1992; Ishikawa, 1995; Jacob et al., 1997; Brandt et al., 2002; Quélo et al., 2007). It is well established that such models provide a good description of the climatological long-range transport. However, the inconvenience in such studies arises from the fact that the simulations of the pollution episodes cannot be easily validated due to the lack of real-time qualitative measurements.

Many simulational studies have been performed in order to predict how the radioactive ^{137}Cs migrated after the accident (e.g. Albeger et al., 1988; Hass et al. 1990; Bonelli et al., 1992; Desiato, 1992; Salvadori et al. 1996; Hatano et al., 1998; Brandt et al., 2002). The primary subject of these studies was emergency evacuation planning over regions within 30 km from the site (called "the exclusion zone"), although most of the results were proven to be inconsistent with the measured data obtained afterwards. Today, more than 25 years after the date of the accident, a better understanding of the fate of radionuclides has been obtained in terms of total deposition. Furthermore, high quality deposition measurements over Europe have become available from the Chernobyl period by the EU Joint Research Centre (JRC) called "the REM database", whereas high resolution maps have been created called "Atlas of caesium deposition on Europe after the Chernobyl accident" (De Cort et al., 1998). These data have being continuously collected by the EU since 1986 in the frame of the REM (Radioactivity Environmental Monitoring) project presenting atmospheric activity concentrations and deposition inventories from European countries, and then used in this paper to validate the model ability to represent the spread and deposition of ^{137}Cs. For the creation of the map (hereafter, referred to as the Atlas), the data have been corrected for radioactive decay to 10 May 1986. A similar map has also been published by Peplow (2006).

Consequently, the main goal of the present work was to study the efficiency of the model described here for the tracer ^{137}Cs using the reported patterns of the Chernobyl releases and transportation over Europe. Therefore, (i) the altitude of the emissions after the episode was considered assuming that the emissions occurred (a) at the surface and (b) at several heights. Moreover, the resulting dispersion and deposition of ^{137}Cs is presented using (ii) the

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regular grid (2.5°×1.27°) and (iii) the zoom-version of the model. Finally, the results of the two versions are evaluated by using two different vertical resolutions: 19 and 39 vertical layers for the regular grid configuration. All the results have been compared with raw data from the REM database. Given the large global risk of human exposure to radiation, especially in areas around reactors in densely populated regions, notably in West Europe and South Asia, where a major reactor accident can expose around 30 million people to radioactive contamination (Lelieveld et al., 2012), a reliable transport model for radioactive substances would be a benefit. The recent decision by Germany (following the Fukushima Daiichi accident in Japan) to phase out its nuclear reactors will reduce the national risk, though a large risk will still remain from the reactors in the neighbouring countries.

2. Global atmospheric transport model

The aerosol module INCA (INteractions between Chemistry and Aerosols) is coupled to the general circulation model (GCM), LMDz, developed at the Laboratoire de Météorologie Dynamique in Paris, and the global vegetation model ORCHIDEE (ORganizing Carbon and Hydrology In Dynamic Ecosystems Environment) (LMDZORINCA) (see also Szopa et al., 2012). Aerosols and gases are treated in the same code to ensure coherence between gas phase chemistry and aerosol dynamics as well as possible interactions between gases and aerosol particles. The simulations using the regular grid described below were performed with a maximum horizontal resolution of 2.5 degrees in longitude and 1.27 degrees in latitude (144×142) (Fig. 1a). However, the GCM also offers the possibility to zoom over specific regions by stretching the grid with the same number of gridboxes (Fig. 1b). In the present study the zoom version was used in Europe obtaining a maximum horizontal resolution of 0.66 degrees in longitude and 0.51 degrees in latitude. On the vertical dimension, the model uses sigma-p coordinates with 19 levels extending from the surface up to about 3.8 hPa corresponding to a vertical resolution of about 300 – 500 m in the planetary boundary layer (first level at 70 m height) and to a resolution of about 2 km at the tropopause (with 7–9 levels located in the stratosphere). Moreover, a vertical resolution of 39 layers has been installed and used extending from the surface up to the mesopause. More information about the parameterisation of wet and dry deposition can be found in Supplementary Material – Methodology.

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Each simulation was carried out for nine months (April to December 1986). Deposition of ^{137}Cs in Europe one month after Chernobyl appeared to be at least two orders of magnitude, or more, lower than the maximum deposition just after the accident, and also that it was fractional (below detection limit) one year later (Kritidis, 1989). Therefore, nine months were sufficient to obtain more than 99% of the ^{137}Cs emitted. LMDZORINCA accounts for emissions, transport (resolved and sub-grid scale), photochemical transformations, and scavenging (dry deposition and washout) of chemical species and aerosols interactively in the GCM. Several versions of the INCA model are currently available depending on the envisaged applications with the chemistry-climate model. The model runs in a nudged mode (using the ERA40 Re-analysis data – 6h wind fields – by the European Centre for Medium-Range Weather Forecasts, ECMWF, 2002) with a relaxation time of 10 days for the regular grid, whereas for the zoom version relaxing to 4.8 days in the center of the zoom and to 10 days outside (Hourdin and Issartel, 2000).

The radioactive tracer ^{137}Cs (half-life = 30.2 years) was inserted as an inert tracer within the model. The behaviour of ^{137}Cs in the atmosphere is strongly related to its chemical form as it may be released in the atmosphere in gaseous form or adsorbed onto particles. Here, it is assumed that mostly ^{137}Cs behaves as an aerosol and as such it is treated in the model. In fact, this is true as it has been reported that over 80% of the ^{137}Cs emitted in the atmosphere during accidental releases is in the form of particulates (Richie and McHenry, 1990; Yoschenko et al., 2006; Sportisse, 2007; Morino et al., 2011; Potiriadis et al., 2011). The partitioning between gaseous form and particles and the size distribution of aerosols strongly affect dry deposition and scavenging.

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3. Emission estimates after the accident

The coordinates of the emissions after the Chernobyl accident in the model were set to 30.083° E longitude and 51.383° N latitude. The precise amount of the emissions after the accident is still laden with uncertainty for the researchers and the local authorities and, typically, an uncertainty of 50 % is used in such analyses (e.g. Albeger et al., 1988; Hass et al. 1990; Brandt et al., 2002). The total source term evaluated by the ex-USSR authorities and published at an IAEA conference in 1986 (Hass et al., 1990) presented a value of 37 (± 50 %) PBq for ^{137}Cs estimated on the basis that all the emitted ^{137}Cs had been deposited in ex-USSR countries only. Nevertheless, a subsequent estimation of the activity of ^{137}Cs emitted after the accident, taking into account the amount of ^{137}Cs deposited in all countries, showed a value more than 2 times higher (85 ± 50% PBq), which is 30% of the total core inventory of ^{137}Cs (280 PBq) (IAEA, 2006). The daily emission percentages (with respect to the total release), the respective activities and the injection height over 19 and 39 vertical layers can be found in Table 1. The major part of the initial emissions from Chernobyl has been estimated to take place at relatively high altitudes. After a few days, the major parts of the emissions were released at lower altitudes below 1.5 km, and in the following days the concentrations were transported over most of Europe with major influences in southern, eastern and central Europe. As a result of the two explosions held during the first day of the accident, the initial large release was due to the mechanical fragmentation of the fuel. It mainly contained the more volatile radionuclides such as noble gases, iodine and some caesium. The second large release in the end of this period was caused by the high temperatures reached in the core melt (Waight et al., 1995).

4. Results and discussion

4.1. Fallout transport over Europe using different model versions

Three separate simulations in the regular grid version of the model were performed, the first one assuming that all the amount of ^{137}Cs was introduced at the site's surface (RG19L(S)), the second following the real emission altitude according to Table 1 spread over 19 layers (RG19L) and the final one over 39 layer resolution following the same emission patterns (RG39L). Moreover, one additional run was performed after installing the zoom-version of the model, stretched over Europe gridded within 19 vertical layers (Z19L) using the emissions denoted in Table 1. The ^{137}Cs activity concentrations in Fig. 2–5 are expressed in Becquerel per m^3 STP, where m^3 STP is a standard cubic meter of air at 273.15 ^{o}K and 1 atm.

The atmospheric activity concentrations of ^{137}Cs from the first run (RG19L(S)) are illustrated in Fig. 2 for the first day of the accident (26th March 1986), for the end of March (30th April 1986), as well as for 5th and 10th May 1986, in order to assess the direction of the radioactive fallout. It is noteworthy that the direction of the radioactive fallout seems not to vary much, mostly affecting the southern countries of Europe and the regions located in

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northern Africa. The atmospheric burden of ^{137}Cs was found to be maximum on the last day of the emission (May 5^{th}) reaching 24 PBq, which corresponds to 28% of the total emitted (Table 2), and then decreased exponentially, presenting an ecological half-life for ^{137}Cs of approximately 3 days (Fig. 6). The ecological half life of ^{137}Cs is defined as the period of time it takes for ^{137}Cs burden to decrease by half, affected by processes others that its radioactive decay (radioactive decay of ^{137}Cs during the 9 months runs was neglected since ^{137}Cs is a long-lived radionuclide presenting a half-life of 30.2 years). Consequently, during the last day of the emissions 28% of ^{137}Cs was still present in the atmosphere, whereas at the end of May the respective rate was 1.1% (1.0 PBq) (Table 2). However, according to the REM database and previous simulations of the accident (e.g. Brandt et al., 2002) the direction depicted by this simulation is inaccurate. This result was expected since the prevailing winds at the surface blow in a very different direction than the ones above.

A closer representation of what happened after the accident is reflected by the second run of the model (RG19L) performed after introducing the known sources of ^{137}Cs at different vertical layers of the model (see Table 1). This simulation indicates that the prevailing advective conditions have spread the radioactive fallout over longer distances than if emission occurred at the surface from the first day of the accident (Fig. 3). At the end of April 1986 the fallout was divided along three axis. The first one was transported to the northern side of Europe, mostly affecting Sweden and Norway and the second part to the western side impacting Central Europe, whereas the final one has a north-eastern direction affecting Russia, Belarus and Ukraine. These results at the end of April concur with the findings of other researchers (Albergel et al., 1988; Brandt et al., 2002; Hass et al., 1990). During the last day of the emissions (5 May 1986), it is evident that the fallout has been distributed over most of Europe. Comparing to the total emission of ^{137}Cs after the accident, 48% (41 PBq) of the ^{137}Cs emitted remained in the atmosphere on 5 May 1986 (Table 2). This is an additional difference between the two runs, since the fallout seems to be deposited more locally if emitted at the surface than at greater heights. The ecological half-life of ^{137}Cs in the atmosphere was estimated by the exponential decrease of the burden and it was found to be almost 6 days (Fig. 6). This differs significantly from the respective ecological half-life estimated during the previous run (RG19L(S), 3 days). Cambray et al. (1987) reported that following the Chernobyl accident, the exponential decline of the ^{137}Cs concentrations indicated a residence half-time of 7 days for ^{137}Cs, which concurs very well with the value found here. The fallout was transferred south-easterly after the end of the emissions, mainly to

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the Middle East, whereas it weakened up to 4 orders of magnitude by the end of May, with only 8% (6.8 PBq) of the total ^{137}Cs emitted still remaining in the atmosphere. The next months the atmospheric burden decreased reaching 0.04 PBq at the end of December, which corresponds to 0.05%, indicating that the vast majority of ^{137}Cs has been deposited by the end of the year 1986 (Table 2). This is in good agreement with the measurements reported elsewhere (e.g. Ballestra et al., 1987; Mattsson and Vasanen, 1988).

Regarding the run performed after installing and using 39 layers in vertical resolution (RG39L) for the same horizontal resolution of the model, there were significant differences in the number of emission vertical levels. In this run the emission distribution in the vertical layers had more points, since the layers were separated by shorter distances between each other. However, the logic of choosing these amounts for each layer (as shown in Table 1) emanated precisely from the previous run (RG19L), in order to achieve similar amounts of ^{137}Cs being emitted from similar altitudes as in the RG19L run. Despite the differences, the tendency of the fallout transport (Fig. 4) was the same as in the RG19L run, since the same ECMWF meteorology files are re-gridded respectively in the vertical plane for both RG19L and RG39L versions. For both resolutions transport occurs to North Europe on the first day of emission (April 26th), whereas the fallout was divided in three components on May 30th, one affecting North Europe (Sweden and Norway), a second one the Central European countries and a third one following a north-eastern direction (across Russia, Belarus and Ukraine). After the last emission date (May 5th) the radioactive plume had been transferred across all Europe. The cyclone observed on May 5th north of the UK influences significantly the wind direction and has been reported by previous investigators in the area. The ecological half-life of ^{137}Cs was estimated to be 9 days, which is higher than for the RG19L run (Fig. 6). In fact, in this run ^{137}Cs was present for longer times in the atmosphere; the burden of ^{137}Cs was estimated at 54 PBq (64%) on May 5th, while the next months decreased exponentially reaching 0.13 PBq (0.15%) at the end of 1986.

The same simulation for the Chernobyl accident was performed, after setting up a zoom-version of the model for 19 vertical layers (Z19L), centred over Europe. The initial transport (April 26th) of the radioactive fallout shows a more pronounced meridional axis than in the previous simulations directed towards West-central Belarus (north), while a much weaker amount of ^{137}Cs (more than two orders of magnitude less) was transferred to Romania and the Black Sea (south) (Fig. 5). The same transport trends have been validated and reported elsewhere (e.g. Brandt et al., 2002, Hass et al., 1990). At the end of April the three

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different directions of the plume (north, west, north-eastern) were apparent and the respective levels were similar to the regular grid runs. On May 5th (last day of the emissions), the plume seems more intense in areas close to the source presenting a more local-based transport, with the fallout extending mostly to the east. ^{137}Cs affects the central and eastern European regions, while it has not been transferred to Spain, Portugal and the North-east African countries yet. Observations support this transport pattern as these countries have reported trace amounts of ^{137}Cs activity concentrations in the air or they were below the respective detection limits. The remaining ^{137}Cs (burden) was maximum on May 5th as it was estimated to be 43 PBq in the atmosphere, which corresponds to 51% of the direct total emission (Table 2). The ecological half-life of ^{137}Cs was also estimated to be almost 6 days (Fig. 6), which is comparable to those estimated by the previous runs and similar to those reported previously for the Chernobyl accident. However, a recent study following the Fukushima NPP accident in Japan showed ecological half-lives of ^{137}Cs to be between 5 and 10 days (Kristiansen et al., 2012). Until May 10th the fallout appeared to follow a southern direction affecting the Middle East, just as in the previous simulations. During the last two thirds of May the radioactive plume over Europe was of the order mBq m^{-3} STP, whereas at the end of the month only 8.2% (7.0 PBq) of the ^{137}Cs emitted still resided into atmosphere. Likewise, ^{137}Cs decreased in the following months and more than 99.9% had been deposited by the end of 1986 (Table 2).

A three dimensional illustration of the 0.15 Bq m^{-3} STP iso-surface of ^{137}Cs on April 28th (12:00 UTC) for 19 (left panel) and 39 vertical layers (right panel) is shown in Fig. 7 as in Brandt et al. (2002). The figure shows what can be seen from the south and ^{137}Cs surface activity concentrations are plotted on the iso-surface. It is noteworthy that some parts of the plume experience vertical transport to higher altitudes. Another important feature here is the fact that the plume is distributed irregularly, both vertically and horizontally, in the 39 layers. It dominates the higher layers of the atmosphere across all Europe, in contrast to the 19 layers run, where the plume ascends mostly near the source. This is actually what Brandt et al. (2002) have proposed: parts of the plume are transported to higher altitudes where the wind direction is opposite to the direction found at lower levels. This wind pattern causes a transport in opposite horizontal directions at different altitudes, i.e. towards northwest at lower altitudes and towards southeast at higher altitudes. The distribution of ^{137}Cs on April 28th extends up to 388 mbars in the 19 layers and up to 74 mbars in the 39. It is obvious that larger amounts of ^{137}Cs have been transferred to higher altitudes in the 39 levels (yellow colors in the iso-surface especially northerly – Fig. 7) resulting to higher residence times of

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^{137}Cs in the atmosphere (see Fig. 6). The Gaussian vertical motion in conjunction with the larger number of the levels spread the radionuclide into higher altitudes, despite our attempts of emitting similar amounts of ^{137}Cs from similar heights in the model.

4.2. Comparison with direct measurements of atmospheric activity concentrations

Data obtained from the four different runs of the Chernobyl accident were compared to real-time measurements of the activity concentrations of ^{137}Cs. For that purpose, the aforementioned REM database was used. The activity concentrations from the database were divided in three parts according to the regions mostly affected by the radioactive fallout on April 30th 1986 (see Fig. 2–5), (a) West-central, (b) North and (c) South Europe. Time series measurements of ^{137}Cs atmospheric concentrations between April and May 1986 can be found in Supplementary Material, Fig. S1 for the regions examined.

In West-central Europe (Germany, France, UK, Belgium, Switzerland, Austria and Netherlands), the trends of ^{137}Cs dispersion in the countries examined showed satisfactory results, in terms of activity concentrations and residence times as well (Supplementary Material, Fig. S1). However, relatively small inaccuracies were observed on some days (e.g. in Austria and Switzerland). The precision of the measurement technique used is not indicated in the database. Data from the REM database have been collected using several different techniques (e.g., direct airborne gamma spectrometry, surface pumping through disc filters followed by gamma spectrometry etc...) and the specific method used at each station is not specified in the database. Therefore, determination recoveries contrast between different methodologies and this might induce additional uncertainties to the results. Regarding the residence time of ^{137}Cs in the countries presented here, the model also shows robustness since, in most of the cases, similar levels were observed. Finally, the ending dates of the fallout, where ^{137}Cs activity concentrations were near the limit of detection (LOD), concur with those of the model.

Similar results were found for the countries of North Europe (Finland, Norway, Sweden and Denmark) with smaller discrepancies (Supplementary Material, Fig. S1). The most apparent were observed for Finland and Norway during the first days of May 1986, where the model underestimates the activity concentrations of ^{137}Cs. However, the patterns of ^{137}Cs

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activity concentrations in all cases were undoubtedly consistent indicating high accuracy for the model. Very similar levels were observed for the starting and the ending point of the radionuclide passage over the countries studied.

Finally, reliable results were obtained for South European countries (ex-Czechoslovakia, Hungary, Italy and Greece) (Supplementary Material, Fig. S1), albeit underestimations over the Italian territory and overestimations over Greece. It is also essential to focus on another source of uncertainty. Even nowadays the exact emissions from the Chernobyl accident are unknown. Therefore, the relatively large discrepancy in the dosages of ^{137}Cs can be explained from discrepancies in the source term or uncertainties in the effective release heights, since the injection altitudes used in the present study are only educated guesses. This seems to be very essential in terms of transport and deposition of ^{137}Cs in certain regions. Another noteworthy point that we should focus on here is what we learn from the results of the RG19L(S) simulation, where surface emissions were assumed. These results differ significantly from measurements and also model-versions where real emission altitudes were used. For instance, no ^{137}Cs was detected in North Europe until the end of May or extreme amount were estimated in South Europe at the start of the same month. This is additional evidence of how the exact height of the emission could affect the subsequent transport of ^{137}Cs and the importance of the uncertainty induced by the source term.

There are several numerical measures that quantify the extent of statistical dependence between pairs of databases. Here, we used the Spearman correlation method (Choi, 1977), which assesses how well the relationship between two variables can be described using a monotonic function. If there are no repeated data values, a perfect Spearman correlation of +1 or −1 occurs when each of the variables is a perfect monotone function of the other. Table 3 shows the respective results of the datasets compared (REM versus RG19L(S), RG19L, RG39L and Z19L, respectively) for the activity concentrations of ^{137}Cs. According to the table, the Spearman correlation coefficient ranged from 0.61 to 0.64 for the runs where the emission altitude was taken into account for 95% confidence level ($p<0.05$), whereas it was 0.21 for the RG19L(S) run; thus the variables are statistically dependent. On the other hand, the data of the simulation with the real emission altitude are also highly dependent presenting coefficients of 0.84. For justification, Kendall rank correlation coefficient, commonly referred as Kendall's tau (τ) coefficient (Christensen, 2005) was also estimated (Table 3). This statistic measures the rank correlation, i.e. the similarity of the orderings of the data when ranked by each of the quantities. It is often used to test a statistical hypothesis in order to establish

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whether two variables may be regarded as statistically dependent. This test is non-parametric, as it does not rely on any assumptions on the distributions of X or Y (X and Y represent the variables under comparison). Under a "null hypothesis" of X and Y being independent, the sampling distribution of τ will have an expected value of zero. Here, the τ values were estimated to be around 0.44 for 95% confidence level (p<0.05); thus, the "null hypothesis" can be rejected and the two datasets are dependent. The data derived from each simulation were very similar with τ coefficients between 0.54 – 0.68. This can also be seen in Fig. 8, which depicts the Box and Whisker plot of the data. The range of the datasets for ^{137}Cs activity concentrations is very similar, whereas the boxes corresponding to 25 – 75% of the values were found at same level, although in some cases the model was found to underestimate. Besides statistics, the average relative biases were also calculated and presented in Supplementary Material, Fig. S1 for each version except the one where surface emissions assumed. Despite the large variation in the biases, they present very satisfactory averages, 9.59, 81.49 and 3.81 for the RG19L, RG39L and Z19L run respectively, which are very good in comparison with previously reported ones for ^{137}Cs activity concentrations of the Chernobyl accident (e.g. –61 in Brandt et al., 2002). The larger positive biases calculated for the 39 levels are a result of the elevation of higher amounts of ^{137}Cs at greater heights (previously discussed in the manuscript) in conjunction with the resulting larger residence times.

Finally, the arrival times of the radioactive fallout of ^{137}Cs were assessed for the four different simulations (RG19L(S), RG19L, RG39L and Z19L) and they were compared to those obtained from the REM database (REM). As arrival time we define the time after the accident it takes for ^{137}Cs to reach the activity concentration of 10^{-4} Bq m^{-3} in a specific location, which is the minimum detected value of the REM database. The results are illustrated in a scatter plot in Fig. 9 for 56 measurement stations in 24 European countries (Ukraine, Russia, Poland, Romania, Hungary, Denmark, Belgium, Finland, Norway, Sweden, Germany, Netherlands, France, Italy, Great Britain, Austria, Switzerland, ex-Czechoslovakia, Turkey, Greece, Ireland, Egypt, Syria and Lebanon). The Pearson linear correlation coefficient was estimated to be 0.65 for the RG19L run, 0.46 for the RG19L and 0.63 for the Z19L, which is considered to be significant. The different vertical resolution resulted in a more rapid transport to the places examined. Moreover, as in the previous comparisons, the respective arrival times of ^{137}Cs estimated from the regular grid run assuming surface

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emissions (RG19L(S)) were not reliable (R^2 = 0.08). The model is able to predict the arrival times of ^{137}Cs for the measurement stations with a good accuracy.

4.3. Deposition of ^{137}Cs in European countries in relation to the Atlas

In this section the atmospheric budget and deposition patterns of ^{137}Cs are assessed taking into consideration the contributions of different removal processes (i.e., particle sedimentation, dry and wet deposition, through large-scale and convective precipitation). The distribution of ^{137}Cs deposited over Europe is shown in Fig. 10 (for the RG19L(S)), whereas in Fig. 11 the Atlas map is illustrated. Fig. 12–14 depict the respective runs with the real emission altitude (RG19L, RG39L and Z19L) for dry (top left panel), wet (top right panel) and total cumulative deposition (lower panel). In these figures, the same scale with the Atlas was used in order to better compare the results. Following the definition given by the International Atomic Energy Agency (IAEA, 2005; 2009), any area with activity larger than 40 kBq m^{-2} is considered to be contaminated (see relevant red scale). Contamination means the presence of a radionuclide on a surface in quantities larger than 40 kBq m^{-2} for beta and gamma emitters (^{137}Cs is a gamma emitter). Since we integrate the deposition over the period after the accident until the end of 1986, the present results represent the cumulated contamination of this radionuclide.

The cumulative dry, wet and total deposition for 1986, estimated assuming that the emissions occurred at the surface (RG19L(S)), are depicted in Fig. 10. These data are presented here in order to certify and record the importance of the altitude of the emission in deposition after accidental releases. As can be seen from Fig. 10 the deposition of ^{137}Cs is largely dependent upon the transport of the atmospheric burden. In the present situation where surface emissions assumed, the deposition was limited locally. It is mainly contingent from the surface southern winds and, following the dominant precipitation, it was deposited in Eastern Europe and the Balkan countries (Russia, Belarus, and Ukraine, ex-Czechoslovakia, Romania, Bulgaria, Greece and ex-Yugoslavia). An example can be given for Kiev (Ukraine), which is the most densely populated city close to the damaged reactor (around 100 km). According to the Atlas (Fig. 11), the deposition of ^{137}Cs in this location appeared to be of the order of 10 – 40 kBq m^{-3}. However, the model predicted a deposition greater than 1480 kBq m^{-3}. That would lead the official authorities to evacuate the city. Therefore, it would be of major importance to know all the information following a major event. It is unexpected what

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would have happened if the official authorities of the ex-USSR evacuated an area of several thousand inhabitants by mistake.

However, the measurements carried out by several accredited laboratories throughout Europe showed that there was transport to many other countries (Fig. 11). A more reliable deposition of ^{137}Cs is reflected by the second run of the model, where ^{137}Cs in the real emission altitude was injected (RG19L). The results are shown in Fig. 12. The transport as well as the dry deposition of ^{137}Cs occurred also throughout Northern European countries especially in the first months after the accident. In addition, the observed precipitation resulted in deposition of higher amounts of ^{137}Cs in specific areas of Sweden and Finland. However, a comparison of the total cumulative deposition of ^{137}Cs simulated by the model to the observed one (De Cort et al., 1998, Fig. 11) showed that the levels of ^{137}Cs deposition are overestimated over Central Europe. The Atlas indicates total deposition inventories of less than 10 kBq m^{-2}, whereas the total deposition inventories estimated in the model were found between 10 and 40 kBq m^{-2}.

The depositional patterns of RG39L simulation of the Chernobyl accident (Fig. 13) are different. The model underestimates the radioactive contamination in the northern countries (in Finland and Sweden), although enhanced depositions were estimated more easterly. Despite these deficiencies the model managed to estimate the increased contamination in the Alpine environment. It has been reported (De Cort et al., 1998) and can be also seen here (Fig. 11) that ^{137}Cs have been deposited in the Alps after the accident, as a results of the intense precipitation. Moreover, the model also predicted effectively the deposition over North Greece.

As expected, the zoom-version of the model (Fig. 14) provides more discrete results of ^{137}Cs deposition over Europe. The relative distribution of ^{137}Cs deposition is similar to the Atlas, although underestimated, whereas some extremely high values of total cumulative deposition appeared in central Europe. The high deposition observed in Sweden is of the same magnitude and also, at the same location as those presented in Atlas. Another good example is the high total cumulative deposition observed in Russia (north-easterly of the Chernobyl NPP) (Carbol et al., 2003), which is predicted by the model accurately (see also Fig. 11). Finally, in Greece, enhanced depositions were observed in continental regions (Kritidis et al., 1990; Kritidis and Florou, 1995), and the model predicted them efficiently (see also Fig. 11). Despite the overestimations observed in Central Europe and underestimations in the highly

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contaminated areas, taking into account the heterogeneity of the direct measurements and the method used to create the Atlas map (inverse distance weighted interpolation method), one could note that the model gives remarkably good results.

4.4. Comparison with depositional observations reported by European countries

Figure S2 in the Supplementary Material shows the location of the measurement stations where measurements of the cumulative total deposition were carried out and presented in the REM-database. Over 4,000 measurements from 20 European countries were used to evaluate and assess the modeling results in terms of the total cumulative deposition of ^{137}Cs. However, no data from Ukraine, Belarus and Russia were available at the EU-JRC. Table 4 shows the respective results of the statistical tests used in order to examine the relevance of the datasets (REM vs RG19L(S), RG19L, RG39L and Z19L, respectively) in contrast to the real-time measurements for ^{137}Cs deposition. The Spearman correlation coefficient was estimated to range from 0.46 to 0.57 with 95% confidence level (p<0.05), whereas the Kendall's tau (τ) rank correlation coefficient was estimated to vary between 0.33 and 0.42, (with 95% confidence level, p<0.05) (Table 4), which shows the dependence between model datasets and observations. In fact, the results obtained from the different model runs (RG19L, RG39L and Z19L) were also contiguous presenting high coefficients (> 0.7). Moreover, Fig. 15 depicts the Box and Whisker plots for the datasets in terms of the total cumulative deposition of ^{137}Cs. There is an obvious trend of the model to underestimate the deposition of ^{137}Cs in the countries examined taking into consideration the boxes corresponding to 25 – 75% of the values, although these ranges were similar. For the comparisons of the model to observations of the deposition of ^{137}Cs, reduced though nevertheless realistic agreement can be claimed, taking into account the inherent uncertainties based on the multitude and the complexity of the simulated removal processes (sedimentation, dry and wet deposition). In most cases there is close coincidence between the modeled and measured deposition inventories of ^{137}Cs, although the simulated deposition fluxes underpredict measured ones by a factor of three in extreme cases. However, the model shows the arrival of high concentrations of radioactively contaminated aerosols at central European countries, and the same transport has been verified by previous models and certified by surface activity concentration measurements.

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Figure S3 in the Supplementary Material gives a more detailed view of the comparisons for each of the 20 European countries. It depicts linear regression scatter plots of the total ^{137}Cs deposition based on individual measurements of each country (REM database) in descending order, in terms of the best linear fitting (1:1 dependence), as well as the respective calculated biases from the comparison with the observations. Given the large heterogeneity of the samples and the 50% uncertainty of the emissions, the model results are in very good agreement with observations. The best performance was achieved for 14 countries (ex-Yugoslavia, Spain, Hungary, Italy, Finland, Sweden, Greece, UK, Netherlands, Switzerland, Belgium, Germany, Norway and France) with correlation coefficients between 0.4 and 0.9, whereas the estimated average bias was –0.81±0.15 for the RG19L run and the Z19L run and lower for the 39 level run (–0.25±0.91). This seems very convenient if compared with other model assessments, which have showed biases around 1.3 for the total deposition of ^{137}Cs (e.g. Brandt et al., 2002). Some discrepancies were observed in countries near the Chernobyl site (Romania, Poland, and ex-Czechoslovakia) and, also underestimations in Denmark, Ireland and Austria, while raw data from Ukraine, Belarus and Russia were unavailable from the public database. An important issue that should be stated here, regarding the data of ^{137}Cs deposition from the REM database, is the fact that these data refer to total deposition of ^{137}Cs over Europe, which means that the respective deposition from global atmospheric weapon testing, as well as other regional releases (e.g. Sellafield in Great Britain, Mayak in Urals, local releases from fuel fabrication etc...) are included in the measurements. We believe that the observed underestimation of the model might be due to the fact that they have been more intensely affected by other releases (e.g. the background of ^{137}Cs in central Europe prior to Chernobyl has been estimated to be greater than 3 kBq m^{-2}). Finally, the precipitation fields were examined in order to assess if the observed difference in the deposition of ^{137}Cs could be owed to non-realistic wet deposition. For this reason, the relative difference between ERA40 ($2.5^o \times 2.5^o$) precipitation and the one used by the LMDZ ($0.66^o \times 0.51^o$) were calculated for 1986. We estimate an average discrepancy of 8% in an area of 700 × 700 km, which increases to 10% in an area of 3000 × 3000 km (all centered over the plant), which is very small and, hence, it is not expected to affect wet processes.

5. Conclusions

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The atmospheric cycle of ^{137}Cs using LMDZORINCA model has been evaluated against real time measurements of ^{137}Cs from the Chernobyl accident in 1986. The model is based on a combination of the aerosol module INCA, the general circulation model LMDz and the global vegetation model ORCHIDEE. The conclusions are based on comparisons with measurements both from the REM database and from the Chernobyl Atlas. Simulations of the Chernobyl accident showed that comprehensive tracer models are powerful tools for estimating the activity concentrations and depositions after accidental scenarios.

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According to the comparison between model and observations, the most sufficient results were obtained when the highest horizontal resolution of the model was used (Z19L run). Specifically, this model version managed to predict the radioactive contamination in most of the regions alike to Atlas. Except for higher coefficients and smaller biases from the comparison with the observations, for every variable examined (e.g. atmospheric activity concentrations, cumulative deposition of ^{137}Cs etc...), a better resolved map similar to Atlas was obtained. However, there is a general trend for underestimation in the deposition, which could be attributed to the prevailing environmental processes and the large uncertainties of the source term, as well as to the background deposition of ^{137}Cs from releases occurred prior to the accident that the model do not account for. The high vertical resolution of 39 levels can be useful only when the exact injection altitude is known. The increased number of levels in the boundary layer resulted in a different dispersion and deposition of ^{137}Cs. When a moderate vertical resolution was used (19 layers in the RG19L run) the results were better. The accurate knowledge of the height of the emission is crucial in order to obtain credible transport and deposition of ^{137}Cs. The resulting transport and deposition, when surface emissions were assumed, appeared to be local event in comparison to what really happened after the accident.

In all realistic situations studied (presenting the real ignition altitude) an ecological half-life of 6 – 9 days was estimated for the global atmospheric burden of ^{137}Cs. In fact, previous modeling studies give global average half-lives of aerosols in the atmosphere on the order of 3–7 days, whereas for the Chernobyl and the recent Fukushima NPP accident a maximum of 10 days has been reported.

In addition, the arrival times of ^{137}Cs in the model in comparison with the observations showed satisfactory correlations (0.46 – 0.65). Expected lack of dependence was estimated when surface emissions were assumed. The model is able to simulate and predict the development of the specific activity fields with high efficiency, although rarely underestimated. This is expected taking into account the uncertainties of the source term, the deposition processes and the heterogeneity in the samples. However, statistical tests applied to the respective datasets proved a likely dependence.

A general conclusion is that the high resolved grid gives results that track closely the observations, especially in the first days of the emissions. This imposes the essential usage of modeling applications as tools for the decision makers, given that the first days of a nuclear accident are very important for life, in terms of addressing the appropriate evacuation criteria for the radiation protection of the population.

There is a critical need for open data policy after accidental releases. It is a pity that no data from all European countries are present in the public section of the REM database. The paper shows the importance of knowing the emission height of the source in such studies and how much it affects the dispersion and deposition of ^{137}Cs. However, only speculations can be made about the real altitude where ^{137}Cs was injected in the atmosphere and therefore, an uncertainty of 50% is always used in the case of Chernobyl. Nowadays, the existence of several modeling tools is able to predict the overall details of the emission after a NPP accident (e.g. using inverse modeling, Stohl et al., 2013). Knowing the exact core inventory by the official authorities or the real emissions during the first days, these dispersion models are able to predict the fate of the radioactive fallout. It is important that such an effort has been made after the recent accident in Japan where the IAEA has created a website with different databases for the Japanese authorities.

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releasing in public the free software Ocean Data View (ODV) and Integrated Data Viewer (IDV), respectively.

References

Albergel, A., Martin, D., Strauss, B. and Gros, J.-M.: The Chernobyl accident: Modeling of dispersion over Europe of the radioactive plume and comparison with air activity measurements, Atmos. Environ., 22, 2431–2444, 1988.

ApSimon H. M., Goddard A. J. H. and Wrigley J.: Long-range atmospheric dispersion of radioisotopses. The MESOS model, Atmos. Environ., 19, 113–125, 1985.

ApSimon H. M., Wilson J. J. N. and Scott P. A.: Assessment of the Chernobyl release in the immediate aftermath of the accident, Nucl. Energy, 26, 295–301, 1987.

Balkanski, Y. J., Jacob, D. J. Arimoto, R., and Kritz, M. A.: Distribution of ^{222}Rn over the North Pacific: Implications for continental influences, J. Atmos. Chem., 14, 353–371, 1992.

Ballestra, S. B., Holm, E., Walton, A., and Whitehead, N. E.: Fallout deposition at Monaco following the Chernobyl accident, J. Environ. Radioact., 5, 391–400, 1987.

Nikolaos Evangeliou 5/13/13 10:50 AM
Deleted: Balkanski, Y. J., Jacob, D. J., Gardner, G. M., and G.K.K. Turekian: Transport and residence times of tropospheric aerosols inferred from a global three-dimensional simulation of ^{210}Pb. J. Geophys. Res., 98, 20573–20586, 1993.

Bonelli, P., Calori, G., and Finzi, G.: A fast long-range transport model for operational use in episode simulation. Application to the Chernobyl accident, Atmos. Environ., 26, 2523–2535, 1992.

Brandt, J., Christensen, J. H., and Frohn, L. M.: Modelling transport and deposition of caesium and iodine from the Chernobyl accident using the DREAM model, Atmos. Chem. Phys., 2, 397–417, 2002.

Cambray, R. S., Cawse, P. A., Garland, J. A., Gibson, J. A. B., Johnson, P., Lewis, G. N. J., Newton, D., Salmon, L. and Wade, B. O.: Observations on radioactivity from the Chernobyl accident, Nucl. Energ. 26, 77–101, 1987.

Nikolaos Evangeliou 5/13/13 10:49 AM
Deleted: Brasseur, G. P., Hauglustaine, D. A., Walters, S., Rasch, P. J., Muller, J.-F., Granier, C., and Tie, X. X.: MOZART, a global chemical transport model for ozone and related chemical tracers: 1. Model description, J. Geophys. Res., 103, 28265–28289, 1998.

Carbol, P., Solatie, D., Erdmann, N., Nylen, T., and Betti, M.: Deposition and distribution of Chernobyl fallout fission products and actinides in a Russian soil profile, J. Environ. Radioact., 68, 27–46, 2003.

Choi, S.C.: Test of equality of dependent correlations, Biometrika, 64, 645–647, 1977.

Christensen, D.: Fast algorithms for the calculation of Kendall's τ, Computation. Stat., 20, 51–62, 2005.

De Cort, M., Dubois, G., Fridman, Sh. D., Germenchuk, M. G., Izrael, Yu. A., Janssens, A., Jones, A. R., Kelly, G. N., Kvasnikova, E. V., Matveenko, I. I., Nazarov, I. M., Pokumeiko, Yu. M., Sitak, V. A., Stukin, E. D., Tabachny, L. Ya., Tsaturov, Yu. S., and Avdyushin, S. I.: Atlas of caesium deposition on Europe after the Chernobyl accident, Luxembourg, Office for Official Publications of the European Communities 1998, ISBN 92-828-3140-X, Catalogue number CG-NA-16-733-29-C. EUR 16733, pp. 1–63, 1998.

Desiato, F.: A long-range dispersion model evaluation study with Chernobyl data, Atmos. Environ., 26, 2805–2820, 1992

Nikolaos Evangeliou 5/13/13 10:53 AM
Deleted: De Fries, R. S., and Townshend, J. R.: NDVI-derived land classification at global scales, Int. J. Remote Sens., 15, 3567– 3586, 1994.

Devell, L., Guntay, S. and Powers, D. A.: The Chernobyl reactor accident source term. Development of a consensus view, NEA/CSNI/R(95)24, OECD Nuclear Energy Agency, Paris, 1996.

Elliassen A.: The OCDE study of long range transport or air pollutants: long-range transport modeling, Atmos. Environ., 12, 479–487, 1978.

Elliassen A. and Saltbones J.: Modelling of long-range transport of sulphur over Europe: a two-year model run and some model experiments, Atmos. Environ., 17, 1457–1473, 1983.

ECMWF: ERA-40, forty-year European re-analysis of the global atmosphere, 2002, available at: http://www.ecmwf.int/products/data/archive/descriptions/e4, 2002.

Hass, H., Memmesheimer, M., Geiss, H., Jakobs, H. J., Laube, M. and Ebel, A.: Simulation of the Chernobyl radioactive cloud over Europe using EURAD model, Atmos. Environ., 24, 673–692, 1990.

Nikolaos Evangeliou 5/13/13 10:47 AM
Deleted: Giorgi, F., and Chameides, W. L.: The rainout parameterization in a photochemical model, J. Geophys. Res., 90, 7872–7880, 1985. ... [2]

Hatano,Y., Hatano, N., Amano, H., Ueno, T., Sukhoruchkin, A.K., and Kazakov, S.V.: Aerosol migration near Chernobyl: long-term data and modeling, Atmos. Environ., 32, 2587–2594, 1998.

Hourdin, F., and Issartel, J. P.: Sub-surface nuclear tests monitoring through the CTBT xenon network, Geophys. Res. Lett., 27, 2245– 2248, 2000.

Nikolaos Evangeliou 5/13/13 11:05 AM
Deleted: Hauglustaine, D. A., Hourdin, F., Walters, S., Jourdain, L., Filiberti, M.-A., Larmarque, J.-F., and Holland, E. A.: Interactive chemistry in the Laboratoire de Météorologie Dynamique general circulation model: description and background tropospheric chemistry evaluation. J. Geophys. Res., 109, D04314, 2004 (doi:10.1029/2003JD003957).

Huneeus, N., Schulz, M., Balkanski, Y., Griesfeller, J., Kinne, S., Prospero, J., Bauer, S., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Fillmore, D., Ghan, S., Ginoux, P., Grini, A., Horowitz, L., Koch, D., Krol, M. C., Landing, W., Liu, X., Mahowald, N., Miller, R., Morcrette, J.-J., Myhre, G., Penner, J., Perlwitz, J., Stier, P., Takemura, T., and Zender, C.: Global dust model intercomparison in AeroCom phase I, Atm. Chem. Phys., 11, 7781–7816, 2011.

International Atomic Energy Agency (IAEA): Regulations for the Safe Transport of Radioactive Material, IAEA Safety Requirements, no. TS-R-1, Vienna, 2005.

International Atomic Energy Agency (IAEA): Environmental consequences of the Chernobyl accident and their remediation: Twenty years of experience. Report of the Chernobyl Forum Expert Group 'Environment', Radiological Assessment Reports Series, Vienna, 2006.

International Atomic Energy Agency (IAEA): Regulations for the Safe Transport of Radioactive Material, IAEA Safety Requirements, no. TS-R-1, Vienna, 2009.

Ishikawa, H.: Evaluation of the effect of horizontal diffusion on the long-range atmospheric transport simulation with Chernobyl data, J. Appl. Meteorol., 34, 1653–1665, 1995.

Jacob, D. J., Prather, M. J., Wofsy, S. C.: Mcelroy M. B. ;Atmospheric distribution of 85Kr simulated with a general circulation modelJGR. J. Geophys. Res. 92 (D6), 6614–6626, 1987.

Jacob, D.J., Prather, M.J., Rasch, P.J., Shia, R.L., Balkanski, Y.J., Beagley, S.R., Bergmann, D.J., Blackshear, W.T., Brown, M., Chiba, M., Chipperfield, M.P., De Grandpré, J., Dignon, J.E., Feichter, J., Genthon, C., Grose, W.L., Kasibhatla, P.S., Köhler, I., Kritz, M.A., Law, K., Penner, J.E., Ramonet, M., Reeves, C.E., Rotman, D.A., Stockwell, D.Z., Van Velthoven, P.F.J., Verver, G., Wild, O., Yang, H., and Zimmermann, P.: Evaluation and intercomparison of global atmospheric transport models using ^{222}Rn and other short-lived tracers, J. Geophys. Res., 102 (D5), 5953–5970, 1997.

Klug, W., Graziani, G., Grippa, G., Pierce, D., and Tassone, C.: Evaluation of long range atmospheric transport models using environmental radioactivity data from the Chernobyl accident, The ATMES Report, Elsevier Applied Science, London and New York, pp. 366, 1992.

Koch, D., Schulz, M., Kinne, S., McNaughton, C., Spackman, J.R.,. Bond, T.C, Balkanski, Y., Bauer, S., Berntsen, T., Boucher, O., Chin, M., Clarke, A., De Luca, N., Dentener, F., Diehl, T., Dubovik, O., Easter, R., Fahey, D.W., Feichter, J., Fillmore, D., Freitag, S., Ghan, S., Ginoux, P., Gong, S., Horowitz, L., Iversen, T., Kirkevåg, A., Klimont, Z., Kondo, Y., Krol, M., Liu, X., Miller, R., Montanaro, V., Moteki, N., Myhre, G., Penner, J.E., Perlwitz, J., Pitari, G., Reddy, S., Sahu, L., Sakamoto, H., Schuster, G., Schwarz, J.P., Seland, Ø., Stier,

Nikolaos Evangeliou 5/13/13 10:55 AM
Deleted: Hwang, W.T., Kim, E.H., Suh, K.S., Jeong, H.J., Han, M.H., and Lee, C.W.: Influence of predictive contamination to agricultural products due to dry and wet processes during an accidental release of radionuclides. Ann. Nucl. Energy, 30, 1457–1471, 2003.

P., Takegawa, N., Takemura, T., Textor, C., van Aardenne, J.A., and Zhao, Y.: Evaluation of black carbon estimations in global aerosol models, Atmos. Chem. Phys., 9, 9001–9026, 2009.

Kornberg, H.A.: The use of element-pairs in radiation hazard assessment, Health Phys., 6, 46–62, 1961.

Kristiansen, N. I., Stohl, A., and Wotawa, G.: Atmospheric removal times of the aerosol-bound radionuclides ^{137}Cs and ^{131}I during the months after the Fukushima Dai-ichi nuclear power plant accident – a constraint for air quality and climate models, Atmos. Chem. Phys. Discuss., 12, 12331–12356, 2012.

Kritidis, P.: Radioactive contamination of the environment. In: Verganelakis, A., Kritidis, P., Economou, L., Papazoglou, I., Papanikolaou, E., Sideris, L., Simopoulos, T. (Eds.), Ourselves and Radioactivity, University of Crete Publications, Heraklion, Greece, pp. 235-270 (in Greek), 1989.

Kritidis, P., and Florou, H.: Environmental study of radioactive caesium in Greek lake fish after the Chernobyl accident, J. Environ. Radioact., 28, 285–293, 1995.

Kritidis, P., Florou, H., and Papanicolaou, E.: Delayed and late impact of the Chernobyl accident on the Greek environment, Radiat. Prot. Dosim., 30, 187–190, 1990.

Lange, R., Dickerson, M. H. and Gudiksen, P. H.: Dose estimates from the Chernobyl accident, Nucl. Technol., 82, 311–323, 1988.

Lee, D.S., Nemitz, E., Fowler, D., and Kingdon, R.D.: Modelling atmospheric mercury transport and deposition across Europe and the UK, Atmos. Environ., 35, 5455–5466, 2001.

Lelieveld, J., Kunkel, D., and Lawrence, M. G.: Global risk of radioactive fallout after major nuclear reactor accidents, Atmos. Chem. Phys., 12, 4245–4258, 2012.

Mattsson, S., and Vesanen, R.: Patterns of Chernobyl fallout in relation to local weather conditions, Environ. Int., 14, 177–180, 1988.

Morino,Y., Ohara, T., and Nishizawa, M.: Atmospheric behavior, deposition, and budget of radioactive materials from the Fukushima Daiichi nuclear power plant in March 2011, Geophys. Res. Lett., 38, L00G11, 2011 (doi:10.1029/2011GL048689).

Nikolaos Evangeliou 5/13/13 10:51 AM

Deleted: Liu, H., Jacob, D. J., Bey, I., and Yantosca, R. M.: Constraints from ^{210}Pb and ^{7}Be on wet deposition and transport in a global three-dimensional chemical transport model driven by assimilated meteorological fields, J. Geophys. Res. 106, 12109–12128, 2001. ... [3]

Olivié, D. J. L., Cariolle, D., Teyssèdre, H., Salas, D., Voldoire, A., Clark, H., Saint-Martin, D., Michou, M., Karcher, F., Balkanski, Y., Gauss, M., Dessens, O., Koffi, B., and Sausen, R.: Modeling the climate impact of road transport, maritime shipping and aviation over the period 1860–2100 with an AOGCM, Atmos. Chem. Phys., 12, 1449–1480, 2012.

Peplow, M.: Counting the dead (special report about the death toll of the Chernobyl accident), Nature, 440, 982–983, 2006.

Piedelievre, J. P., Musson-Genon, L. and Bompay, F.: MEDIA - An Eulerian model for atmospheric dispersion: First validation on the Chernobyl release, J. Appl. Meteorol., 29, 1205–1220, 1990.

Potiriadis, C., Kolovou, M., Clouvas, A., and Xanthos, S.: Environmental radioactivity measurements in Greece following the Fukushima Daichi nuclear accident, Radiat. Prot. Dosim., 150, 441–447, 2011.

Prather, M., McElroy, M., Wofsy, S., Russell, G., Rind, D.: Chemistry of the global troposphere: Fluorocarbons as tracers of air motion, J. Geophys. Res., 92 (D6), 6579–6613, 1987.

Quélo, D., Krysta, M., Bocquet, M., Isnard, O.,Minier, Y., Sportisse, B.: Validation of the Polyphemus platform on the ETEX, Chernobyl and Algeciras cases, Atmos. Environ., 41, 5300–5315, 2007.

Ritchie, J.C., and McHenry, J.R.: Application of radioactive fallout cesium-137 for measuring soil erosion and sediment accumulation rates and patterns: A review, J. Environ. Qual., 19, 215–233, 1990.

Salvadori, G., Ratti, S.P., and Belli, G.: Modelling the Chernobyl radioactive fallout (II): A multifractal approach in some European countries, Chemosphere, 33, 2359–2371, 1996.

Sportisse, B.: A review of parameterizations for modelling dry deposition and scavenging of radionuclides, Atmos. Environ., 41, 2683–2698, 2007.

Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and Petzold, A.: The Aerosol-Climate Model ECHAM5-HAM, Atmos. Chem. Phys., 5, 1125–1156, 2005.

Nikolaos Evangeliou 6/4/13 11:55 AM
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Nikolaos Evangeliou 5/13/13 10:48 AM
Deleted: Seinfeld, J. H., and Pandis S. N.: Atmospheric Chemistry and Physics, John Wiley, New York, 1998.

Stohl, A., Seibert, P., Wotawa, G., Arnold, D., Burkhart, J. F., Eckhardt, S., Tapia, C., Vargas, A., Yasunari, T. J.: Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Daiichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition, Atmos. Chem. Phys., 12, 2313−2343, 2012.

Szopa, S., Balkanski, Y., Cozic, A., Deandreis, C., Dufresne, J.-L., Hauglustaine, D., Lathiere, J., Schulz, M., Vuolo, R., Yan, N., Marchand, M., Bekki, S., Cugnet, D., Idelkadi, A., Denvil, S., Foujols, M.-A., Caubel, A., and Fortems-Cheiney, A.: Aerosol and Ozone changes as forcing for Climate Evolution between 1850 and 2100, Clim. Dyn., in press doi:10.1007/s00382-012-1408-y, 2012.

Valkama, I., Salonoja, M., Toivonen, H., Lahtinen, J., and Pollanen, R.: Transport of radioactive gases and particles in the Chernobyl accident: Comparison of environmental measurements and dispersion calculations, International Atomic Energy Agency, International Symposium on Environmental Impact of Radioactive Releases, Vienna, Austria, 8–12 May 1995, IAEA-SM-339/69, pp. 10, 1995.

Nikolaos Evangeliou 5/13/13 10:47 AM
Deleted: Valkama, I. and Pollanen, R.: Transport of radioactive materials in convective clouds, In: Proceedings of the 14th International Conference on Nucleation and Atmospheric Aerosols, Helsinki, August 26-30, 1996, (Eds) Kulmala, M. and Wagner, P. E., p. 4, 1996.

Waight, P., Métivier, H., Jacob, P., Souchkevitch, G., Viktorsson, C., Bennett, B., Hance, R., Kumazawa, S., Kusumi, S., Bouville, A., Sinnaeve, J., Ilari, O., and Lazo, E.: Chernobyl Ten Years On. Radiological and Health Impact. An Assessment by the NEA Committee on 10 Radiation Protection and Public Health, November 1995, OECD Nuclear Agency, http://www.nea.fr/, pp. 74, 1995.

Woodhead, D.S.: Levels of radioactivity in the marine environment and the dose commitment to the marine organisms, I.A.E.A. – S.M. 158/31, Vienna, 1973.

Nikolaos Evangeliou 5/13/13 10:52 AM
Deleted: Walcek, C. J., Brost, R. A.,. Chang, J. S, and Wesely, M. L.: SO_2, sulfate and HNO_3 deposition velocities computed using regional landuse and meteorological data, Atmos. Environ., 20, 949– 964, 1986. ... [4]

Yoschenko, V. I., Kashparov, V.A., Protsak, V.P., Lundin, S.M., Levchuk, S.E., Kadygrib, A.M., Zvarich, S.I., Khomutinin, Yu.V., Maloshtan, I.M., Lanshin, V.P., Kovtun, M.V., Tschiersch, J.: Resuspension and redistribution of radionuclides during grassland and forest fires in the Chernobyl exclusion zone: part I. Fire experiments, J. Environ. Radioact., 86, 143–163, 2006.

Table 1. Daily emissions of ^{137}Cs in PBq after the Chernobyl accident in 1986 (according to Brandt et al. (2002)) and relative vertical distribution in the model for 19 and 39 layers.

Mid-point in 19 Layers (m)	Layer thickness (m)	April 26th (24 %)	April 27th (8 %)	April 28th (6.8 %)	April 29th (5.2 %)	April 30th (4 %)	May 1st (4 %)	May 2nd (8 %)	May 3rd (10 %)	May 4th (14 %)	May 5th (16)
140	45-235	-	-	1.450	1.000	0.850	0.850	1.700	2.150	3.050	3.500
360	236-486	-	0.335	2.900	2.000	1.700	1.700	3.400	4.300	6.100	7.000
690	486-895	-	3.735	1.450	1.000	0.850	0.850	1.700	2.150	3.050	3.500
1200	896-1505	14.050	2.700	-	-	-	-	-	-	-	-
1900	1506-2295	5.050	-	-	-	-	-	-	-	-	-
2900	2296-3505	1.000	-	-	-	-	-	-	-	-	-
Mid-point in 39 Layers (m)	**Layer thickness (m)**	**April 26th (24 %)**	**April 27th (8 %)**	**April 28th (6.8 %)**	**April 29th (5.2 %)**	**April 30th (4 %)**	**May 1st (4 %)**	**May 2nd (8 %)**	**May 3rd (10 %)**	**May 4th (14 %)**	**May 5th (16)**
208	0-243	-	-	0.580	0.400	0.340	0.340	0.680	0.860	1.220	1.400
278	244-326	-	-	0.725	0.500	0.424	0.424	0.850	1.075	1.526	1.751
372	327-443	-	0.134	1.305	0.900	0.765	0.765	1.531	1.934	2.745	3.150

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508	444-609	-	0.134	1.160	0.800	0.680	0.680	1.361	1.719	2.440	2.800
700	610-841	-	1.748	1.232	0.850	0.722	0.722	1.446	1.828	2.593	2.975
963	842-1154	1.405	1.681	0.652	0.450	0.382	0.382	0.765	0.968	1.373	1.576
1309	1155-1567	11.24	1.994	0.145	0.100	0.085	0.085	0.170	0.215	0.305	0.350
1754	1568-2095	3.425	1.080	-	-	-	-	-	-	-	-
2308	2096-2755	2.020	-	-	-	-	-	-	-	-	-
2982	2756-3562	1.610	-	-	-	-	-	-	-	-	-
3779	3563-4522	0.400	-	-	-	-	-	-	-	-	-
TOTAL		**20.10**	**6.771**	**5.800**	**3.999**	**3.398**	**3.398**	**6.803**	**8.600**	**12.201**	**14.001**

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Table 2. Atmospheric burden of ^{137}Cs in PBq (with respect to the total emission of 85 PBq) estimated from the different model-versions used for the Chernobyl simulation. RG19L(S) denotes the simulation in the regular grid (144x142) assuming surface emissions, RG19L the regular grid with the real emission height, RG39L the regular grid with a 39 layer vertical resolution and Z19L the zoom-version over 19 vertical layers.

	April 26th	April 30th	May 5th	May 31st	June 30th	July 31st	August 31st	September 30th	October 31st	November 30th	December 31st
RG19L(S)	8.4	13	24	1.0	0.1	0.04	0.02	0.02	0.01	< 0.01	< 0.01
RG19L	11	27	41	6.8	1.1	0.19	0.09	0.07	0.5	0.04	0.04
RG39L	10	29	54	7.1	1.3	0.28	0.18	0.16	0.14	0.13	0.13
Z19L	12	28	43	7.0	1.2	0.25	0.13	0.10	0.08	0.07	0.06

Table 3. Comparison of the activity concentrations of ^{137}Cs from the Chernobyl accident between the REM database and the different model versions used (RG19L(S), RG19L, RG39L and Z19L). Spearman Rank Order and Kendall Tau correlations (R^2) between the datasets (N = 711) for 95 % confidence level (p<0.05).

	Spearman Rank Order correlation						Kendall Tau correlation				
	REM	**RG19L(S)**	**RG19L**	**RG39L**	**Z19L**		**REM**	**RG19L(S)**	**RG19L**	**RG39L**	**Z19L**
REM	**1.00**	0.21	0.64	0.61	0.62	**REM**	**1.00**	0.16	0.45	0.43	0.44
RG19L(S)	0.21	**1.00**	0.11	0.20	0.15	**RG19L(S)**	0.16	**1.00**	0.08	0.15	0.11
RG19L	0.64	0.11	**1.00**	0.84	0.84	**RG19L**	0.45	0.08	**1.00**	0.67	0.68
RG39L	0.61	0.20	0.84	**1.00**	0.72	**RG39L**	0.43	0.15	0.67	**1.00**	0.54
Z19L	0.62	0.15	0.84	0.72	**1.00**	**Z19L**	0.44	0.11	0.68	0.54	**1.00**

Table 4. Comparison of the total cumulative deposition of ^{137}Cs from the Chernobyl accident between the REM database and the different model versions used (RG19L(S), RG19L, RG39L and Z19L). Spearman Rank Order and Kendall Tau correlations (R^2) between the datasets (N = 4266) for 95 % confidence level ($p<0.05$).

	Spearman Rank Order correlation						Kendall Tau correlation				
	REM	**RG19L(S)**	**RG19L**	**RG39L**	**Z19L**		**REM**	**RG19L(S)**	**RG19L**	**RG39L**	**Z19L**
REM	**1.00**	0.08	0.52	0.46	0.57	**REM**	**1.00**	0.05	0.39	0.33	0.42
RG19L(S)	0.08	**1.00**	0.23	0.37	0.25	**RG19L(S)**	0.05	**1.00**	0.16	0.26	0.18
RG19L	0.52	0.23	**1.00**	0.89	0.93	**RG19L**	0.39	0.16	**1.00**	0.74	0.81
RG39L	0.46	0.37	0.89	**1.00**	0.87	**RG39L**	0.33	0.26	0.74	**1.00**	0.71
Z19L	0.57	0.25	0.93	0.87	**1.00**	**Z19L**	0.42	0.18	0.81	0.71	**1.00**

FIGURE CAPTIONS

Fig. 1. a. 144×142 (points) regular grid of the GCM used for the simulations of the Chernobyl accident using 19 and 39 sigma-p vertical layers, **b.** 144×142 grid "stretched" over Europe (zoom-version) for 19 vertical layers.

Fig. 2. Daily mean surface ^{137}Cs activity concentrations (in Bq m^{-3} STP) from the Chernobyl accident. Model with regular grid and 19 vertical levels assuming surface emissions (RG19L(S)). The figures show the situation during the first day (26th April 1986), at the end of April (30th April 1986), the last day of the emissions (5th May 1986) and in 10th May 1986.

Fig. 3. Daily mean surface ^{137}Cs activity concentrations (in Bq m^{-3} STP) from the Chernobyl accident. Model with regular grid and 19 vertical levels and injection at the real emission height (Table 1) (RG19L). The figures show the situation during the first day (26th April 1986), at the end of April (30th April 1986), the last day of the emissions (5th May 1986) and in 10th May 1986.

Fig. 4. Daily mean surface ^{137}Cs activity concentrations (in Bq m^{-3} STP) from the Chernobyl accident. Model with regular grid and 39 vertical levels and injection at the real emission height (Table 1) (RG39L). The figures show the situation during the first day (26th April 1986), at the end of April (30th April 1986), the last day of the emissions (5th May 1986) and in 10th May 1986.

Fig. 5. Daily mean surface ^{137}Cs activity concentrations (in Bq m^{-3} STP) from the Chernobyl accident. Model with regular grid stretched over Europe and 19 vertical levels and injection at the real emission height (Table 1) (Z19L). The figures show the situation during the first day (26th April 1986), at the end of April (30th April 1986), the last day of the emissions (5th May 1986) and in 10th May 1986.

Fig. 6. Exponential decrease of the atmospheric burden of ^{137}Cs (in PBq) for the 4 different simulations of the Chernobyl accident (RG19L(S), RG19L, RG39L and Z19L). This graph was used in order to estimate the ecological half-lives of ^{137}Cs in the atmosphere. R^2 is the correlation coefficient of the exponential fitting that the burden of ^{137}Cs in the atmosphere follows.

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Fig. 7. A three-dimensional mapping of the 0.15 Bq m^{-3} STP iso-surface of ^{137}Cs on the third day after the Chernobyl accident (April 28th, 12:00UTC) for 19 (left panel) and 39 vertical levels (right panel). Surface activity concentrations in Bq m^{-3} STP are plotted on the iso-surface with the darker color indicating high concentrations and the lighter lower ones).

Fig. 8. Box & Whisker plots of the surface activity concentrations of ^{137}Cs obtained from the REM database and from the simulations using all the available versions of the model. The plot depicts the smallest observation (sample minimum), lower quartile, median, upper quartile and the largest observation (sample maximum) (N = 711).

Fig. 9. Estimation of the arrival times of the radioactive fallout of ^{137}Cs after simulation using all model versions (RG19L(S), RG19L, RG39L and Z19L) and comparison with the respective ones obtained from the REM database. The data correspond to time-series measurements from 56 sampling points across Europe.

Fig. 10. Cumulative dry, wet and total deposition of ^{137}Cs (kBq m^{-2}) from the day of the accident (26th April 1986) until the end of 1986. Results of the simulation using the regular grid of the model for 19 vertical layers and accounting for source emissions to occur at the surface (RG19L(S)).

Fig. 11. The Atlas map depicting the total cumulative deposition of ^{137}Cs throughout Europe as a result of the Chernobyl accident and nuclear weapon testing from all available data of the REM database corrected for radioactive decay to 10 May 1986. The map has been published in the "Atlas of caesium deposition on Europe after the Chernobyl accident" (De Cort et al., 1998).

Fig. 12. Cumulative dry, wet and total deposition of ^{137}Cs (kBq m^{-2}) from the day of the accident (26th April 1986) until the end of 1986. Results of the simulation using the regular grid of the model for 19 vertical layers and accounting for the altitude of the emissions with respect to Table 1 (RG19L).

Fig. 13. Cumulative dry, wet and total deposition of ^{137}Cs (kBq m^{-2}) from the day of the accident (26th April 1986) until the end of 1986. Results of the simulation using the regular grid of the model for 39 vertical layers and accounting for the altitude of the emissions with respect to Table 1 (RG39L).

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Fig. 14. Cumulative dry, wet and total deposition of ^{137}Cs (kBq m^{-2}) from the day of the accident (26th April 1986) until the end of 1986. Results of the simulation using the zoom-version of the model for 19 vertical layers and accounting for the altitude of the emissions with respect to Table 1.

Fig. 15. Box & Whisker plots of the cumulative total deposition of ^{137}Cs obtained from the REM database and from the simulations using all the available versions of the model (RG19L(S), RG19L, RG39L and Z19L for 1986. The plot depicts the smallest observation (sample minimum), lower quartile, median, upper quartile and the largest observation (sample maximum) (N = 4266).

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Simulations of the transport and deposition of ^{137}Cs over Europe after the Chernobyl NPP accident: Influence of varying emission-altitude and model horizontal and vertical resolution

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SUPPLEMENTARY MATERIAL

Methodology – Parameterisation of deposition (wet and dry), diffusion and convection

The deposition of an atmospheric constituent over a given terrain depends on the local wind speed, the sensible heat, the land-use data (vegetation type, water, soil, etc...), the characteristics of the compound (as e.g. whether it is in gaseous or in particulate form, or both) and on precipitation (cloud physics). Deposition is defined as the amount of the air pollutant (in both forms), which is transferred to the earth's surface by wet and dry removal processes. It is a time dependent process and varies with meteorological conditions and precipitation. In the present study, ^{137}Cs was assumed to be in particulate form (it is treated as a sub-micronic aerosol in accumulation mode following a lognormal distribution), when released from the nuclear power plant, although the particle size was uncertain, ranging between 0.01 and 50 µm, (Valkama and Pollanen, 1996), and as sub-micronic aerosol in wet scavenging.

The LMDz general circulation model distinguishes between stratiform and convective precipitation. The wet scavenging is calculated in INCA for both types of precipitation separately and parameterized as a first-order loss process (Giorgi and Chameides, 1985):

$$\frac{d}{dt}C_g = -\beta C_g \qquad (1)$$

where C_g is the gas phase concentration of the considered species and β the scavenging coefficient (1/s). The scavenging associated with large-scale stratiform precipitation is calculated adopting the falling raindrop approach and calculating the amount of gas removed by the drop falling through each model layer located below the cloud level (Seinfeld and Pandis, 1998). The increase of the aqueous phase concentration C_{aq}^m of an irreversibly scavenged gas in a droplet originating from level m and falling through a model layer i (where layer i < layer m) can be estimated by a mass balance between the rate of increase of the mass of species in the droplet and the rate of transfer of species to the drop (Seinfeld and Pandis, 1998):

$$\left[\frac{d}{dt}C_{aq}^m\right]_i = \frac{6K_c}{D_p}C_g^i \qquad (2)$$

where C_g^i is the gas phase concentration in layer i encountered by the drop originating from level m, D_p is the rain droplet diameter fixed to a constant value of 3×10^{-3} m in this version of INCA, and K_c the mass transfer coefficient (m s^{-1}). The mass transfer is calculated until equilibrium of the dissolved gas is eventually reached in the falling drop. K_c is calculated with the relation given by Brasseur et al. (1998). In this relation, we assume a constant value for the drop terminal velocity, we assume that rainout is suppressed at temperatures below 258 ^{o}K.

The scavenging by convective precipitation is calculated as part of the upward convective mass flux on the basis of a modified version of the scheme proposed by Balkanski et al. (1993). On the basis of this formulation and on the basis of equation (1), it can be derived, for the scavenging coefficient associated with convective precipitation,

$$\beta^{cv} = -fF_u\frac{g}{p} \qquad (3)$$

where f is the fraction of soluble gas removed from the gas phase, F_u the upward convective mass flux diagnosed by the GCM (kg m^{-2} s^{-1}), p the pressure and g the gravity constant.

As in the study by Liu et al. (2001), we assume that in the convective column,

$$f = 1 - e^{-a\Delta z} \qquad (4)$$

where Δz (m) is the height in the convective tower calculated from the cloud base. The scavenging efficiency a (m^{-1}) is calculated as the ratio of the rate constant for conversion of cloud water to precipitation (C_{pr}) and the updraft velocity w. On the basis of Mari et al. (2000) and Liu et al. (2001), we adopt C_{pr} = 5×10^{-3} s^{-1}, w = 10 m s^{-1} leading to a = 5×10^{-4} m^{-1}.

The dry deposition of ^{137}Cs was computed using the analogy of surface resistance. The deposition velocity is defined as the inverse of the sum of an aerodynamic resistance and a surface resistance placed in series (Balkanski et al., 1993). They are calculated by the following equation:

$$v_d = \frac{1}{R_a+R_b+R_c} \qquad (5)$$

where R_a, R_b and R_c (s m^{-1}) are the aerodynamical, quasi-laminar, and surface resistances, respectively. R_a and R_b are calculated on the basis of Walcek et al. (1986). The surface resistances are determined for all species included in LMDZORINCA according to their Henry law equilibrium constant and reactivity factor for oxidation of biological substances. The surface resistances are calculated using the vegetation map classification from De Fries and Townshend (1994) interpolated to the model grid and redistributed into the classification proposed by Wesely (1989). The lower and upper canopy resistances (including stomata, mesophyll, and cuticle resistances) as well as ground resistances are all parameterised according to Wesely (1989). Meteorological variables needed to calculate R_a, R_b and R_c (including temperature, specific humidity, wind speed, precipitation, snow cover, and solar radiation at the surface) are provided by the GCM at each time step. The deposition velocities used in the model for that restricted study area (Europe, 10° W – 80° E, 25° – 75° N) ranged between 0.05 cm s^{-1} over ocean and 0.2 cm s^{-1} over land depending on the period of study. These values are within the range of deposition velocities used in such studies, e.g.0.04 – 0.5 cm s^{-1} (Sehmel, 1980), 0.31 (Slinn and Slinne, 1980), 0.1 cm s^{-1} (Hanna, 1991), 0.05 cm s^{-1} (Maryon et al., 1992) 0.1 – 0.5 cm s^{-1} (Klug et al., 1992) and 0.1 cm s^{-1} (Hwang et al., 2003).

Following Laval et al. (1981), the turbulent eddy diffusivity is computed as:

$$K_z = max\left(l^2 \left\|\frac{\partial \vec{V}}{\partial z}\right\| \sqrt{1 - \frac{R_i}{R_{i_c}}}, K_{min}\right) \qquad (6)$$

where the mixing length l is prescribed as $l = l_0(\frac{p}{p_s})^2$ with $l_0 = 35\,m$, $R_i = ((\frac{g}{\vartheta})(\frac{\partial \vartheta}{\partial z}))/\left\|\frac{\partial \vec{V}}{\partial z}\right\|^2$ is the local Richardson number and $R_{i_c}(= 0.4)$ is a critical Richardson number. Over continents and ice, the value of the minimum diffusivity, $K_{min} = 10^{-7} m^2 s^{-1}$, was tuned in order to get the right strength for the polar inversion (Hourdin et al., 2006). Over oceans, in order to obtain a satisfactory contrast between trade wind cumuli and strato-cumuli on the eastern borders of basins, a diffusion coefficient K_z is first computed with a very small minimum diffusivity $K_{min} = 10^{-10} m^2 s^{-1}$. A second ad-hoc (and generally stronger) diffusivity, $K_z = \xi l^2$ with $\xi = 0.002\, s^{-1}$, is used if the temperature inversion at the boundary layer top is weak (in practice if the maximum value of the vertical gradient of potential temperature, $-\frac{\partial \vartheta}{\partial z}$, is greater than 0.02 K/Pa). The first coefficient is mainly active in the subsidence regions, especially on the East side of oceanic basins. The second one produces smaller (in fact too small) cloud covers in regions of trade wind cumuli. Surface fluxes are computed using parameters (roughness length, albedo, temperature, humidity etc.) adapted to each surface type. For each atmospheric column, vertical diffusion is applied independently for each subsurface, and the resulting tendencies are averaged (Hourdin et al., 2006).

The parameterization of convection in the model is reported by Hourdin et al. (2006). With respect to the Tiedtke's scheme used in previous versions, the Emanuel's scheme improves the representation of the Hadley–Walker circulation, with a relatively stronger and deeper large-scale ascent over continents, and suppresses the unrealistic patterns of strong rainfall over tropical oceans. Thanks to the regime-sorted framework, originally proposed by Bony et al. (2004) to analyse the cloud radiative forcing and sensitivity, these differences were attributed to intrinsic differences in the vertical distribution of the convective heating and to the lack of self-inhibition by precipitating downdraughts for the Tiedtke's scheme. The combined use of velocity (or z-weighted) potential to characterize the large-scale circulation on the one hand, and regime-sorted approach on the other, appears as a promising framework to work on the validation and improvement of the physical content of atmospheric general circulation models.

References

Balkanski, Y. J., Jacob, D. J., Gardner, G. M., and G.K.K. Turekian: Transport and residence times of tropospheric aerosols inferred from a global three-dimensional simulation of ^{210}Pb. J. Geophys. Res., 98, 20573–20586, 1993.

Brasseur, G. P., Hauglustaine, D. A., Walters, S., Rasch, P. J., Muller, J.-F., Granier, C., and Tie, X. X.: MOZART, a global chemical transport model for ozone and related chemical tracers: 1. Model description, J. Geophys. Res., 103, 28265–28289, 1998.

Bony, S., Dufresne. J,-L,, Le Treut, H., Morcrette, J.-J., Senior, C.: On dynamic and thermodynamic components of cloud changes, Clim. Dyn., 22, 71–86, 2004.

De Fries, R. S., and Townshend, J. R.: NDVI-derived land classification at global scales, Int. J. Remote Sens., 15, 3567– 3586, 1994.

Giorgi, F., and Chameides, W. L.: The rainout parameterization in a photochemical model, J. Geophys. Res., 90, 7872–7880, 1985.

Hanna, S. R., Gifford, F. A., and Yamartino, R. J.: Long range radioactive plume transport simulation Model/Code – Phase I. USNRC Division of Contracts and Property Management, Contract Administration Branch, P-902, Washington, DC 20555. Technical report, pp. 121, 1991.

Hourdin, F., Musat, I., Bony, S., Braconnot, P., Codron, F., Dufresne, J.-L., Fairhead, L., Filiberti, M.-A., Friedlingstein, P., Grandpeix, J.-Y., Krinner, G., LeVan, P., Li, Z.-X., Lott, F.: The LMDZ4 general circulation model: climate performance and sensitivity to parametrized physics with emphasis on tropical convection, Clim Dyn, 27, 787–813, 2006.

Hwang, W.T., Kim, E.H., Suh, K.S., Jeong, H.J., Han, M.H., and Lee, C.W.: Influence of predictive contamination to agricultural products due to dry and wet processes during an accidental release of radionuclides. Ann. Nucl. Energy, 30, 1457–1471, 2003.

Klug, W., Graziani, G., Grippa, G., Pierce, D., and Tassone, C.: Evaluation of long range atmospheric transport models using environmental radioactivity data from the Chernobyl accident, The ATMES Report, Elsevier Applied Science, London and New York, pp. 366, 1992.

Laval, K., Sadourny, R., Serafini, Y.: Land surface processes in a simplified general circulation model, Geophys. Astrophys. Fluid Dyn., 17, 129–150, 1981.

Liu, H., Jacob, D. J., Bey, I., and Yantosca, R. M.: Constraints from ^{210}Pb and ^{7}Be on wet deposition and transport in a global three-dimensional chemical transport model driven by assimilated meteorological fields, J. Geophys. Res. 106, 12109–12128, 2001.

Mari, C., D. J. Jacob, and P. Bechtold: Transport and scavenging of soluble gases in a deep convective cloud, J. Geophys. Res., 105, 22255–22268, 2000.

Maryon, R.H., Smith, F.B., Conwy, B.J., Goddard, D.M.: The UK nuclear accident response model (NAME). Progr. Nucl. Energ., 26 (2), 85–104, 1992.

Sehmel, G.A.: Particle and gas deposition: a review. Atmos. Environ., 14, 983–1011, 1980.

Seinfeld, J. H., and Pandis S. N.: Atmospheric Chemistry and Physics, John Wiley, New York, 1998.

Slinn, S.A., Slinne, W.G.N.: Prediction for particle deposition on natural waters. Atmos. Environ., 14, 1013–1016, 1980.

Valkama, I. and Pollanen, R.: Transport of radioactive materials in convective clouds, In: Proceedings of the 14th International Conference on Nucleation and Atmospheric Aerosols, Helsinki, August 26-30, 1996, (Eds) Kulmala, M. and Wagner, P. E., p. 4, 1996.

Walcek, C. J., Brost, R. A., Chang, J. S, and Wesely, M. L.: SO_2, sulfate and HNO_3 deposition velocities computed using regional landuse and meteorological data, Atmos. Environ., 20, 949– 964, 1986.

Wesely, M. L.: Parametrization of surface resistances to gaseous dry deposition in regional-scale numerical models, Atmos. Environ., 23, 1293–1304, 1989.

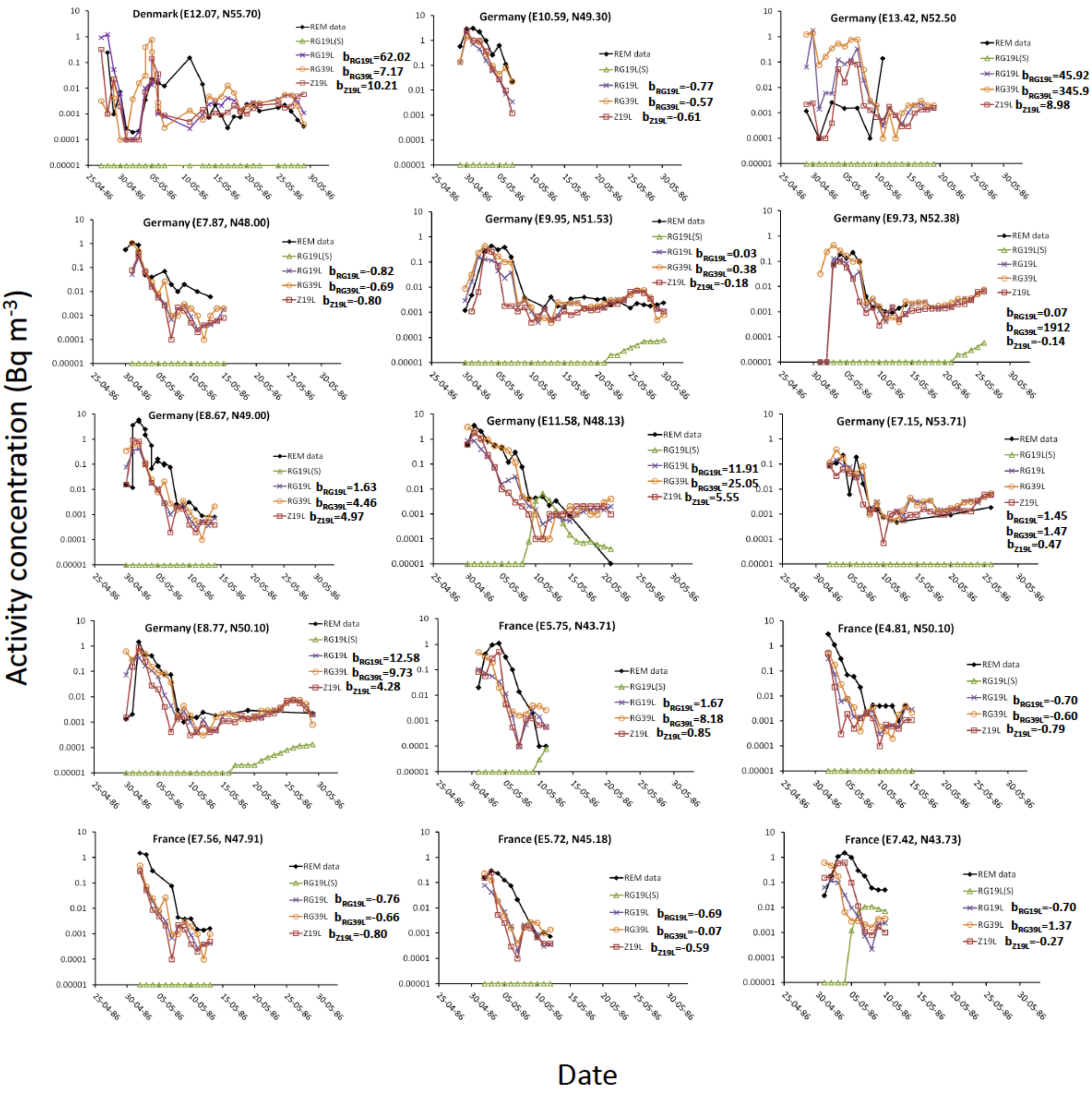
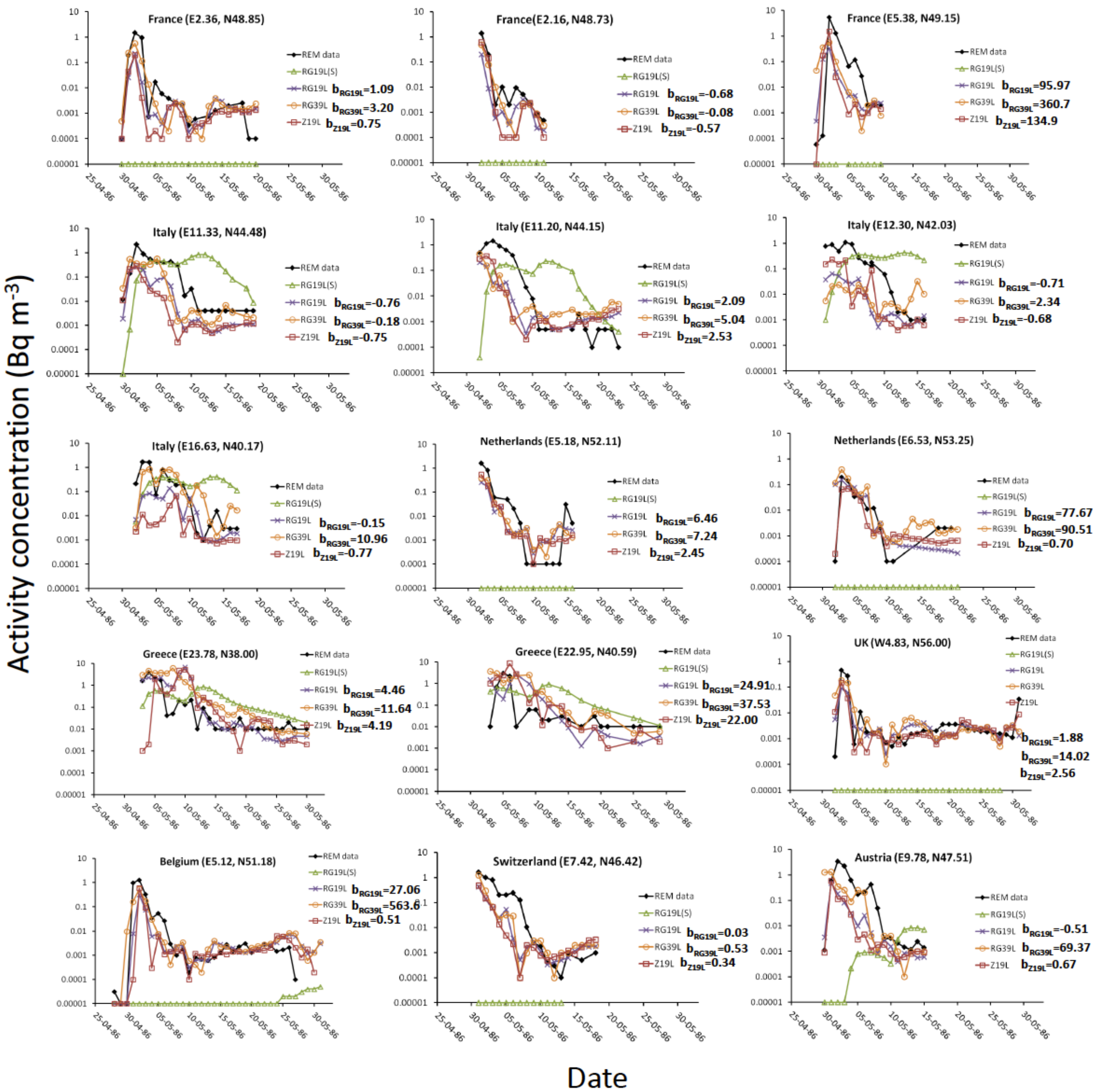


Figure S1. Comparison of the ^{137}Cs surface activity concentrations estimated by all model versions with observations reported in the "REM database" for the Chernobyl accident. The data are available in the website of EU Joint Research Centre in Ispra, Italy. They were examined according to the 3 different directions of the fallout (north, west, south-eastern) on 30th April 1986. The estimated biases are also shown for all the runs (b_RG19L, b_RG39L and b_Z19L) except the one where surface emissions assumed.



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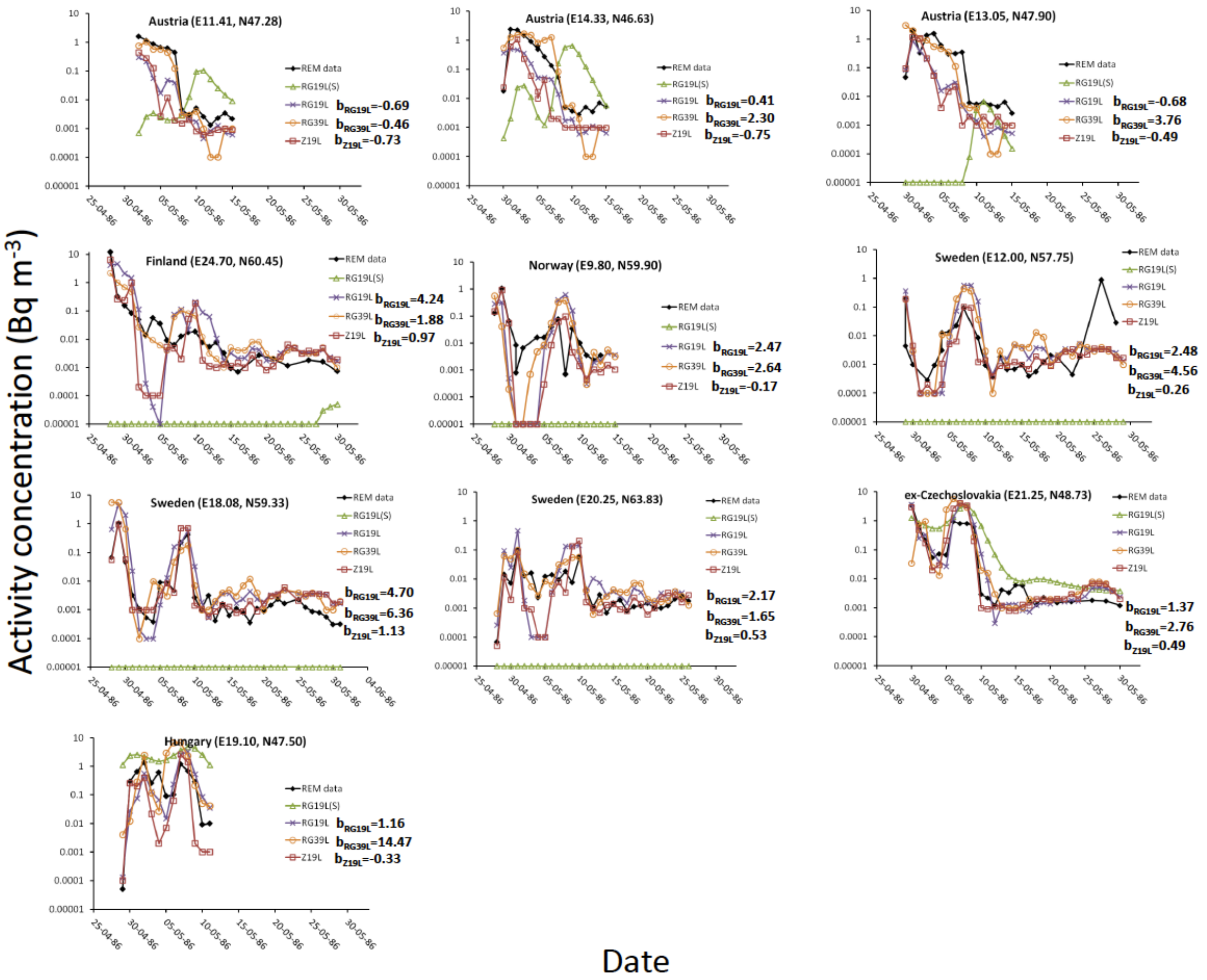

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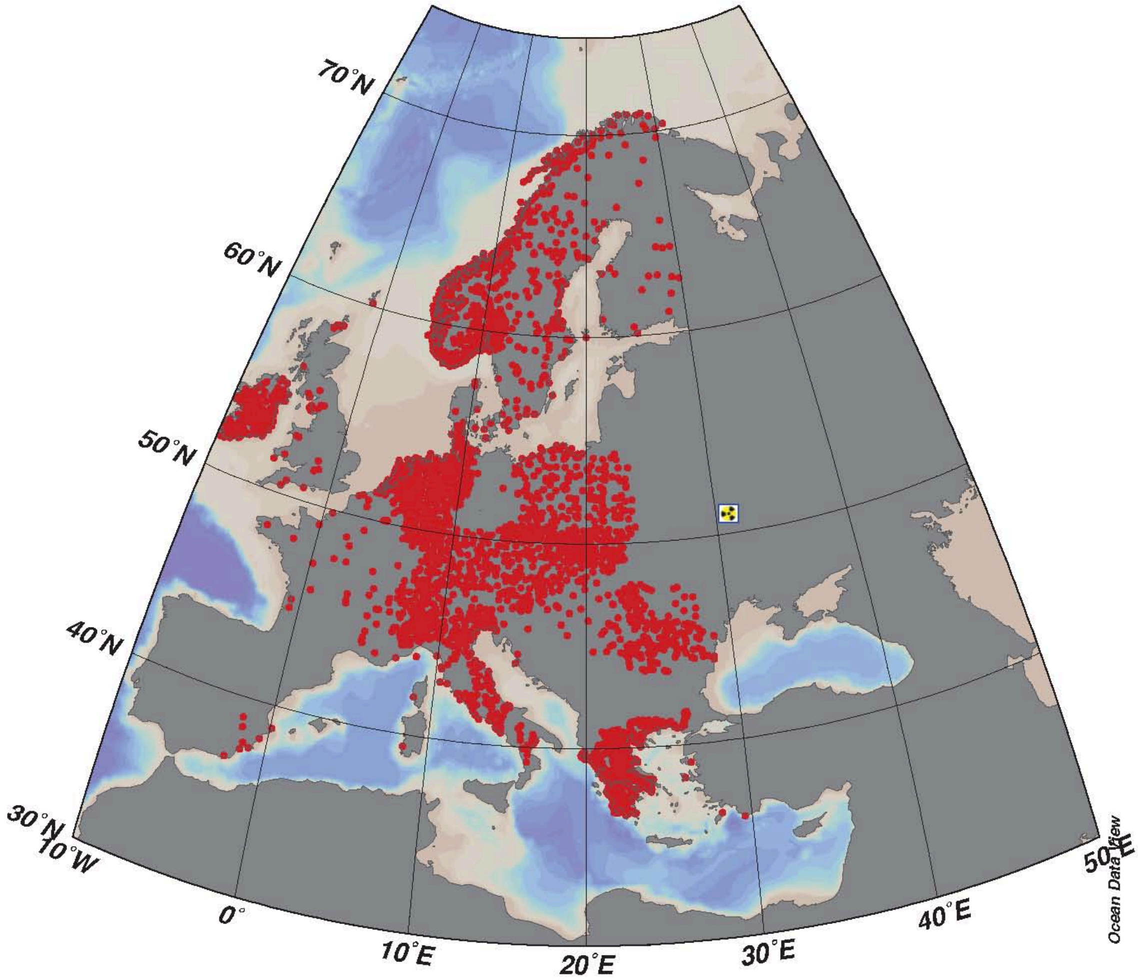

Figure S2. Location of the sampling stations (N = 4266), where measurements of the total deposition of ^{137}Cs after the Chernobyl accident (from the REM database) were carried out. The data were used for the validation of the total deposition of ^{137}Cs resulting from the simulations of the Chernobyl accident using all available versions of the model.

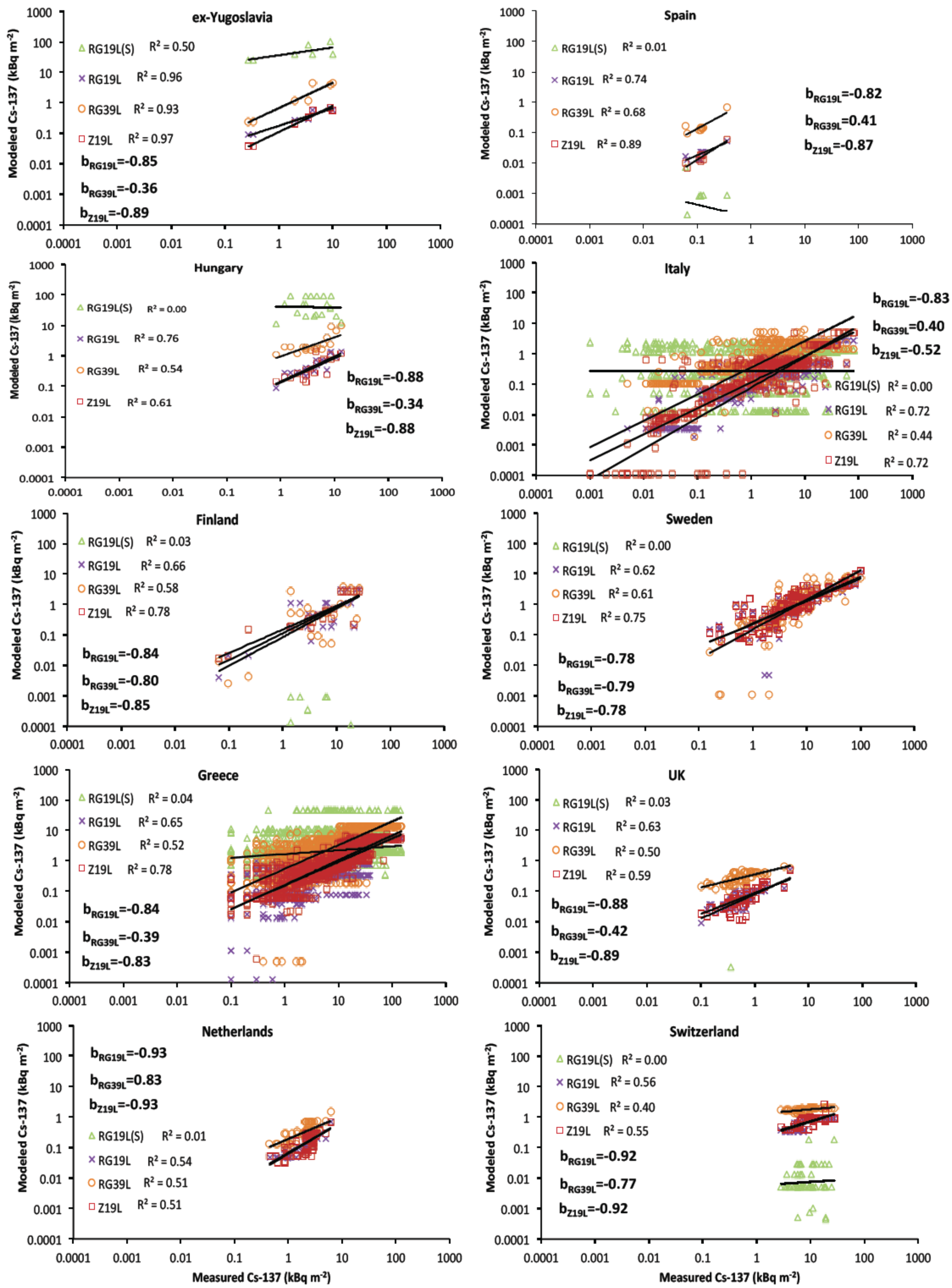

Figure S3. Linear regression scatter plots of the cumulative deposition of ^{137}Cs in 20 European countries from the simulations of all model versions (Modeled Cs-137) and the REM database (Measured Cs-137). The plots are presented in descending order from the best to the worst linear fitting (1 :1). The estimated biases are also shown for all the runs (b_RG19L, b_RG39L and b_Z19L) except for the one where surface emissions assumed.

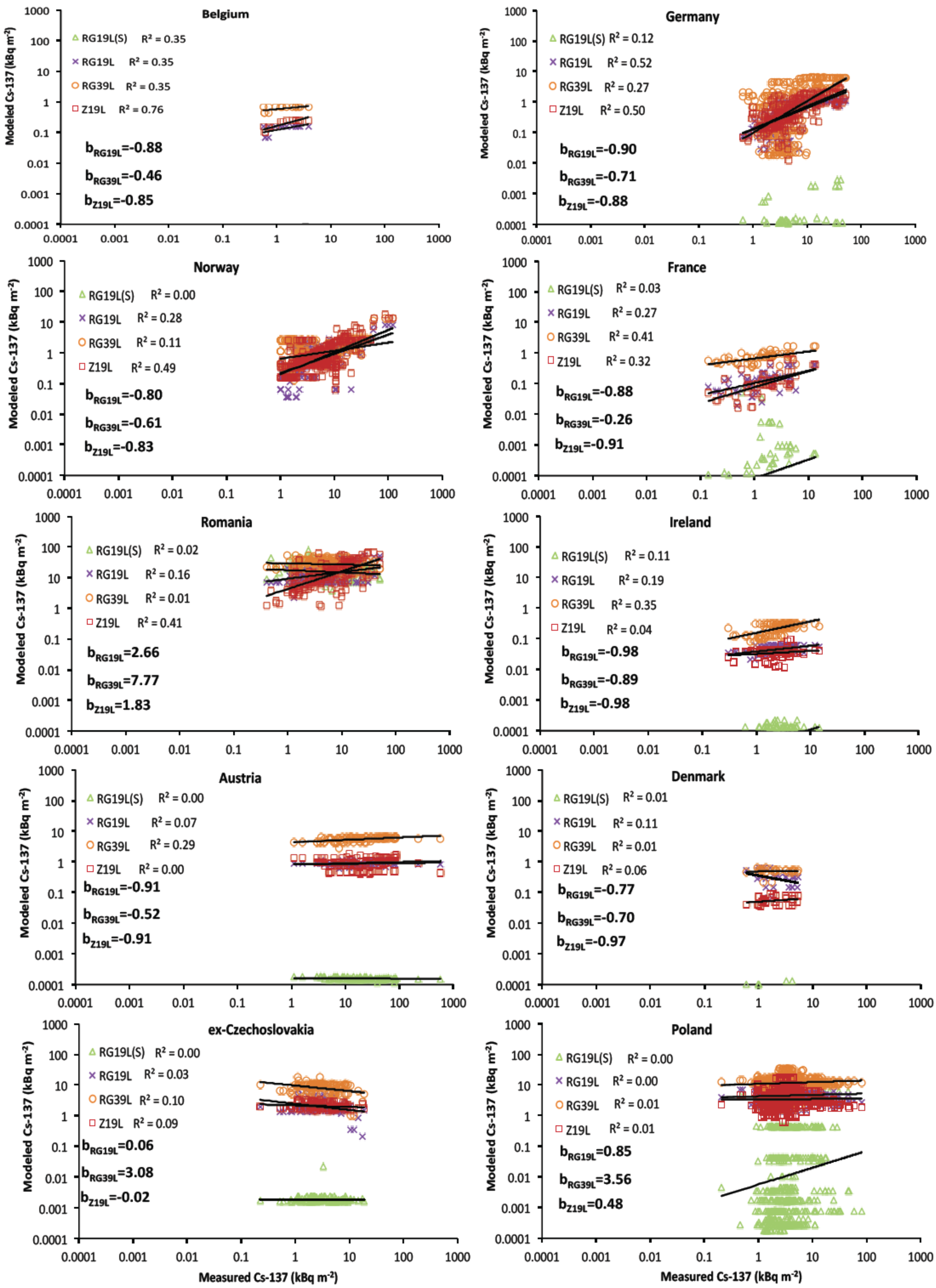

Figure S3. Continued.